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Water quality in relation to rainfall and land-use using California mussels

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WATER QUALITY IN RELATION TO RAINFALL AND LAND-USE USING CALIFORNIA MUSSELS

A THESIS SUBMITTED TO

THE FACULTY OF THE DIVISION OF SOCIAL SCIENCES

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DEPARTMENT OF ENVIRONMENTAL STUDIES

BY

TIANNE M. LARSON

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ABSTRACT

WATER QUALITY IN RELATION TO RAINFALL AND LAND-USE USING CALIFORNIA MUSSELS

By Tianne M. Larson

The purpose of this thesis is to investigate the long-term bioaccumulation trends of copper, lead, silver, total chlordane, total dichlorodiphenyltrichloroethane (DDT), and total polychlorinated biphenyls (PCBs) in relation to rainfall and land-use in the California mussel, *Mytilus californianus.*

From 1977 to 2000, the State Mussel Watch Program (SMWP) collected mussels at 547 sampling sites. This thesis focused on 42 sites. Regressions were performed to determine trends using the statistical package, SPSS. Temporal analysis revealed that 25% of the sites showed significant decreases in lead concentrations, 29% decreased in silver, 25% increased in copper, 50% decreased in total chlordane, 50% decreased in total DDT, 42% decreased in total PCBs and 29% increased in total PCBs.

Using a stepwise multivariate regression statistical technique, developed land uses were associated with concentrations of heavy metals and agricultural land uses were associated with increases in agricultural chemicals. This thesis concludes with recommendations for further study.

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INTRODUCTION

Purpose of Study

The purpose of this study is to investigate the long-term bioaccumulation trends of copper, lead, silver, total chlordane, total DDT, and total PCBs in relation to rainfall and land-use in the California mussel, *Mytilus californianus* (hereinafter referred to as the California mussel or "mussel"). Silver, copper, lead, total chlordane, total DDT, and total PCBs are assessed for significant changes in their concentrations resulting from rainfall precipitation and land-use. The conclusions of the study will aid in the design of future work designed to monitor the complexities of human impacts that affect the concentrations of chemical contaminants in the marine environment.

Background

Coastal and estuarine regions are critical to our nation's economy and human quality of life. Yet, these ecosystems are among our most damaged environments. Chemicals from land-based sources such as urban runoff, agricultural runoff, and treated wastewater, introduce potentially toxic pollutants to coastal waters. These pollutants are heavily affecting nationwide coastal areas. In California, near-shore marine waters have been negatively impacted over recent years as more Californians have moved to coastal regions.

Since European settlement in the mid-1800s, the California coast has been irreversibly altered by heavy agricultural, industrial, and urban development. According to Kreissman (1991), in the past 200 years, California has lost 89% of its riparian woodland and 80% of its coastal wetlands because of irrigation, drainage activities, flood

control, grading, paving and construction. Further impacts can be expected as the population of California continues to increase. The 2000 U.S. census revealed that California's population increased from 15.7 million in 1960 to 33.9 million in 2000 and is projected to reach 49.3 million by 2025 (U.S. Census, 2000).

As the United States' population increases, people are recognizing that long-term economic well-being cannot be sustained without safeguarding natural resources. In 1972, the Clean Water Act (CWA) was passed to re-establish and preserve the chemical, physical, and biological integrity of the Nation's waters. Since 1972, remarkable water quality improvements have occurred as a result of increased control over point-source pollution. Now, non-point sources (NPS) of pollution, such as urban and agricultural run-off, pose the most serious threat to our water quality.

Shellfish bans, beach closures, accidental spills of oil and toxic chemicals, losses of wetland habitat, and fish kills are the types of events that have aroused public concern over the quality and sustainability of the marine environment (O'Connor, 1998). The sources and pathways of contaminants that cause these problems are vast. Most scientists and policy-makers now recognize that protecting our marine environment and changing coastal management can alleviate many of the negative health and quality of life effects on humans caused by coastal pollution. Effective coastal-zone management decisions require accurate data on the ecosystem changes caused by human activities, such as agricultural drainage, waste inputs, commercial trade and water diversion verses those caused by natural change such as weather events (i.e. El Niño Southern Oscillation and La Niña events).

Government officials, academic researchers, and citizens need dependable information on coastal environmental conditions to manage coastal areas for sustainable use. Current, detailed scientific information on how our natural resources respond over time to toxic trace elements and organic contaminants is needed in order to sustain an ecologically diverse and productive ecosystem. It is necessary to obtain data on past and present chemical contaminant inputs to determine if environmental conditions are improving, deteriorating or remaining the same. Regular monitoring of chemical contaminants in our coastal areas can indicate changes in our ecosystem and sources of pollution (Phillips, 1997; A. Munoz-Barbosa *et al.*, 2000).

The California coast, both water and land, is a complex and sensitive ecosystem facing numerous problems. We must gain the necessary knowledge of how the problems we face on land (i.e. wetland retention, water conservation, lack of water sources, etc.) affect water quality (agricultural runoff, waste treatment, overuse of water, etc.) (Herrmann *et al, 1991*). Herrmann states in his article how important long-term monitoring and research is in providing a context for developing more meaningful hypotheses. He emphasizes that more effort should be made to use results from current long-term studies for the development and calibration of complex models of aquatic and watershed ecosystems.

Implications

As human-induced pressures on our coastal areas intensify, new approaches to environmental monitoring will be necessary in order to balance economic development with environmental preservation. The results of this study provide valuable data on

contaminant concentrations, which expands the existing body of literature and aids planners of environmental policy and monitoring programs. The State Water Quality Control Board will be able to use the data found in this study to help identify specific watersheds impacted by chemical contaminants, establish thresholds of wastewater effluent, and impose regulatory restrictions. The major effect of human intervention in regards to pesticide pollution occurred in the 1970s. As a result, trends occurring in the 1980s are smaller and more easily masked by natural effects on interannual variation. Therefore, monitoring studies will need to be done on a continual basis to accrue more data and allow for more conclusive, statistically powerful data analysis.

RELATED RESEARCH

Mussels and chemical contamination are the subject of numerous studies worldwide. The following discussion explores the various agencies and their important empirical research in this field, which focuses on mussel contaminant reports and methods used in evaluating these studies.

Environmental Monitoring Organizations

Several management programs have been monitoring for trace metals and organic compounds in the California marine environment. Specifically, the impact of metal and organic contaminants on the ecology of the marine environment has been the focus of several programs, including the National Oceanic Atmospheric Administration (NOAA), the Regional Monitoring Program (RMP), and the State Mussel Watch Program (SMWP). However, the available data collected by these programs are often non-comparable and inconclusive, creating the need for a more synergistic monitoring approach (Olsen *et al.*, 1999).

The SMWP, founded in 1977 by the State Water Resources Control Board (SWRCB), was established to prepare a uniform statewide approach in detecting and monitoring the concentration of toxic substances in California's bays, harbors, and estuaries as measured in the California mussel, a bivalve commonly found along the California coast. Numerous studies have been conducted in relation to metal

contamination of mussels due to their ability to concentrate contaminants and their extensive distribution and abundance.

The SWRCB is the state agency responsible for implementing and overseeing water quality programs in California. This agency provides funding under an ongoing interagency agreement with the California Department of Fish and Game (CDFG) to collect and analyze bivalve and sediment samples statewide. The Regional Water Quality Control Board (RWQCB), the SWRCB, and the EPA use the information collected to identify water bodies affected by toxic chemicals and to classify water quality issues (Rasmussen, 1996). Through the implementation of a uniform statewide program, the SWRCB has been able to evaluate the effectiveness of water quality programs used in the protection of coastal waters and their native inhabitants.

Several studies conducted by NOAA's National Status and Trends Mussel Watch Program (NS&T) found that there were no apparent temporal trends in the majority of the studied chemicals at the majority of the sites. However, where trends did exist, chemical concentrations of trace metals (i.e. arsenic, cadmium, copper, mercury, nickel, lead, selenium, and zinc) and organic compounds (i.e. total chlordane, total DDT, dieldrin, total PCBs, polycyclic aromatic hydrocarbons (PAHs) and tributyltin (TBT)) were typically decreasing overtime (O'Connor, 1992, 1994, 1996; O'Connor *et al.*, 1994).

Despite the decreases found by O'Connor (1996), a few California sites showed increasing trends for arsenic, cadmium, copper, mercury, nickel, zinc, and PAHs. In the 1998 report, U.S. Long-term Coastal Contaminant Temporal Trends Determined from Mollusk Monitoring Programs, 1965-1993, Lauenstein *et al.* combined contaminant data

from four large-scale monitoring programs to determine long-term trends in U.S. coastal waters. Decreasing national trends for cadmium, lead, silver, total PCBs, total DDT and its metabolites, and dieldrin were found. Decreasing and increasing trends were approximately equal for copper, except for in California where increasing trends were evident at five of the 36 sites. The long-term study found no trends for PAHs, nickel or zinc concentrations.

Stephenson and Leonard (1994) studied California coastal sites and found trends of decreasing concentrations of silver and lead, as well as increasing concentrations of copper from 1977 through 1990. They suggested that the decreasing trends in silver and lead were a result of the decrease in mass emissions from wastewater treatment plants, improved reclamation efforts, and the phase-out of leaded gasoline. Stephenson and Leonard believed that the increase in copper levels could be a result of the increase in boat traffic and the use of anti-fouling paints containing copper on boats. Their findings are similar to those of Lauenstein *et al.* (1990) and O'Connor (1992) who conducted their studies in coastal and estuarine regions of the U.S. for the NS&T Mussel Watch Program. O'Connor detected changes in the concentrations of elements in 13% of the 5-year data set, whereas, Stephenson and Leonard showed changes in 40% of their 14-year data set.

Lauenstein *et al.* (1990) found the test sites in Southern California to have the majority of the decreasing lead trends. Lauenstein *et al.* (1990) stated the following: "A national decrease of lead concentration in aquatic organisms is expected because of the elimination of lead in gasoline in the U.S. in 1990. Transportation emissions, which accounted for over 80 percent of total lead emissions in 1970, declined by more than 98

percent from 1970 to 1980 while emissions from industrial and stationary fuel combustion sources, which are the second and third largest sources of airborne lead, respectively, declined by more than 90 percent (USEPA, 1990)."

Daskalakis *et al.* (1997) also found decreasing trends in silver concentrations in mussels. Daskalakis believed that such decreases were a reflection of the improved recovery of silver from solutions used in the photography industry, which accounts for over 50% of silver use in the United States. The photography industry began decreasing its use of silver in the late 1970's (Lauenstein and Daskalakis, 1998).

Studies show decreasing trends for DDT and its metabolites on a national scale along all coasts (Lauenstein and Daskalakis, 1998, Stephenson, Martin, and Tjeerdema, 1995, Wade and Sericano, 1998). Since the use of DDT was banned in the United States in 1972 and was greatly curtailed world wide, decreased concentrations of DDT in the environment are to be expected (Lauenstein and Daskalakis, 1998). However, due to an estimated half-life of 10-20 years, the decrease of DDT has been sluggish (Sericano *et al.*, 1990).

Other mussel studies have found decreasing trends for PCBs along the U.S. estuarine and coastal environments (Lauenstein and Daskalakis, 1998 and Stephenson, Martin, and Tjeerdema, 1995).

Bioavailability of Chemicals

Complex interactions between organisms, food, and geochemistry control the biological availability of chemicals in mussels (Salazar *et al.*, 2000). Their digestive enzymes, gut amino acids, gut retention time, contaminant loading and the available food

are a few of the intricate interactions that contribute to uncertainty in predicting bioavailability of chemicals in bivalves (Salazar *et al.*, 2000). The mechanisms of absorption across the gut and the importance of dietary exposure pathways are not adequately understood.

Mussels are known to have the ability to keenly detect copper and close their valves to avoid exposure for periods up to two weeks and still survive. Although mussels are commonly used for bioaccumulation testing, they are not recommended for toxicity testing because of their ability to close their valves to avoid chemical exposure.

Phytoplankton are frequently the principal food source for filter-feeding and deposit-feeding organisms and can increase the bioavailability of chemicals in mussels as nutrient-rich particles with associated chemicals are ingested. Salazar and Salazar (2000) stated that filter feeders often compete for food sources, including floating particulates such as bacteria, algae, and sediment; all of which are capable of becoming a chemical pathway of exposure.

Importance of Local and Climatic Influences

Kim *et al.* (2001) has observed that the variability in chemical concentration in mussels is controlled by complex local or watershed-dependent factors, such as land-use and freshwater inflow, and by larger-scale climatic factors, such as temperature and precipitation. Urban runoff transported through storm drains and direct wastewater discharge of toxic compounds from industrial and municipal treatment plants into aquatic

ecosystems represents an increasing environmental problem potentially affecting both marine life and human health (Ciccotelli, Crippa, and Colombo, 1998).

As part of NOAA's NS&T program, oysters were collected along the Gulf of Mexico from 1986 through 1993. In this study, Kim *et al.* (2001) looked at the geographic trends in contaminant concentration and found that the body burden of most metal contaminants were controlled by local or watershed-dependent factors, such as land use and freshwater inflow. Most metals were not affected spatially or temporally by climatic factors. However, as an exception to this general finding, the body burden of metals (As, Cd, Hg, Ni, and Se) seemed to be significantly influenced by temperature and precipitation. In contrast to the metals, Kim *et al.* (2001) found that the temporal trends of most organic contaminants and biological attributes were strongly influenced by climate. However, the spatial trends were not influenced by climate.

Studies show that chemical concentrations increase with increasing rainfall. Wilson *et al.* (1992) identified the influence of geographic patterns of temperature and precipitation.

A study done in 1999 by Gary Ishikawa, in Moss Landing, California, showed a significant correlation between rainfall and DDT in the tissues of mussels at the SMWP's Moss Landing site. At the time of writing, the SMWP concentration data for the Moss Landing site was the only mussel watch site that had been analyzed with rainfall data (Ishikawa, personal communication).

In another study, after 6 days of heavy rainfall (9.6 cm) in January 1995, the Pajaro Valley Water Management Agency (PVWMA) reported increased DDT

concentrations with increased river flow. The concentration of DDT increased from $0.13\mu g l^{-1}$ when the river flow was $8.3 m^3 s^{-1}$ to $1.1\mu g l^{-1}$ when the river flow increased to $79.6 m^3 s^{-1}$ (Hunt *et al.*, 1999).

Hunt *et al.* (1999) found toxicity values to be significantly correlated with rainfall and river flow in the Pajaro River estuarine system in central California. Their study showed increasing rainfall and river flow were correlated with decreasing resident mysid crustaceans (*N. mercedis*) survival rates. They observed high water flow due to rainfall facilitated the transport of pollutants, primarily organochlorine pesticides, from urbanized and agricultural areas throughout the watershed. They concluded that the upper river and the agricultural drainage ditches that served as tributaries were more significant than freshwater sloughs as sources of toxic runoff into the Pajaro River estuary. This study highlighted the importance of land-use in contaminant research.

Land-use Studies

Land-use change, primarily increases in urban/impervious areas, appears to have major short and long-term impacts on water resources from increased downstream flooding to decreased groundwater supplies. It is suggested that storm water from urban areas is a leading cause of water quality impairment.

Studying an entire watershed from 1973 through 1991, Bhaduri *et al.* (2000) found that an 18% increase in urban or impervious areas yielded an estimated 80% increase in annual average runoff volume. The annual average loads for copper, lead, and

zinc increased by more than 50%. They also found that nitrogen and phosphorus loads decreased by 15%, largely due to the loss of agricultural land.

The following studies have shown that elevated levels of inorganic and organic chemicals may be related to the growth in population and industrialization of an area. The studies conducted by Sericano (1993), Craig *et al.* (1989), and Wilson *et al.* (1992) related the distribution of selected chemicals to particular watershed uses such as urbanization and industrialization. Cantillo and O'Connor (1992) found that chemical concentrations at NS&T sites were strongly influenced by the sites' proximity to population centers. Martin and Castle (1984) found that chlordane showed higher concentrations in urban areas.

Influence of Wastewater Treatment Facilities on Mussels

Alexander and Young (1976) observed that wastewater discharges have the capacity to influence areas that are located a significant distance away. They reported that silver concentrations in resident California Mussels reflected a pattern radiating away from the southern California sewage discharges located at Whites Point outfall at Palos Verdes and Santa Monica Bay in Los Angles County. Bruland *et al.* (1974) found the Los Angeles municipal outfall discharges, which contain lead and zinc, resulted in suspended particulates collected near Santa Catalina Island, which is located approximately fifteen kilometers from the White Point outfall.

In another study, Goldberg *et al.* (1983) found a correlation between major municipal waste outfalls and elevated concentrations of silver and lead. Brown *et al.* (1985) found "gradients of chlorinated hydrocarbons (principally DDT and PCB

compounds) in livers of scorpion fish roughly proportional to distance from the White Point outfall" in Palos Verdes, California. In respect to the concentration of silver, mussels harvested from Point Dume in Santa Monica Bay showed a strong mark of sewage discharge from the Whites Point outfall. Brown *et al.* (1985) also discovered that sediments near the Whites Point outfall had high levels of DDTs, PCBs and metals.

Martin *et al.* (1988) showed that silver, zinc and DDT most often originate from municipal wastewater discharges in southern California. They reported that transplanted mussels in outfall dilution areas rapidly absorb and bioconcentrate silver. They found a strong correlation between zinc and silver outfall discharge and mussel concentrations. They also found the concentrations of silver in the tissues of mussels surrounding outfall sites to be higher than those used as reference or from non-influenced sites. The silver concentrations in the mussels from the Royal Palms mussel watch site in southern California were inversely proportional to the distance from the major municipal plants.

Despite the fact that numerous studies have established that municipal wastes are important contributors of waste-derived contaminants in mussel tissue, many measurable improvements have been made in the quality of wastewater effluent due to more aggressive source control and upgraded wastewater treatment plants. With respect to the chemical contamination in mussels, most studies in marine environments have involved only the quantity of chemicals found, not the sources and distributions of the pollutants.

This thesis provides additional data on the temporal and spatial distributions of copper, lead, silver, total chlordane, total DDTs, and total PCBs found in California mussels along California's coastline and identifies the probable sources of these

pollutants. This research adds to the existing knowledge of anthropocentric impacts on mussels and the marine environment at the selected mussel watch stations by investigating the relationship between land-use, rainfall, and contaminant loading to receiving water bodies and California mussels.

RESEARCH OBJECTIVES

The primary objective of this study is to see if there are any possible relationships between the different land-uses, quantity of rainfall, and chemical runoff into the receiving water bodies and the absorption of these chemicals into the tissue of the California mussels along the California coast. The specific objectives and hypotheses are as follows:

Objective One: Identify temporal trends in contaminant concentrations and distribution of trace metals (copper, lead, and silver) and organic chemicals (total chlordane, total DDT, and total PCBs) in the tissue of the California mussel along the California coast from 1990-2000 to determine whether possible trends are consistent with trends found in previous studies (Stephenson and Leonard, 1994; Stephenson, Martin, and Tjeerdema, 1995). Specifically, the null and alternative hypotheses are:

H_0: Additional data from (1990-2000) does not change significant trends in lead and silver levels at the sites.

H_1: Given the more recent data over the past decade (1990-2000), there has continued to be a significant reduction of lead and silver in sites that previously showed significant decreases (Stephenson and Leonard 1994).

H_0: Additional data from (1990-2000) does not change significant trends in copper levels at the sites.

H_1: Given the more recent data over the past decade (1990-2000), there has continued to be a significant increase of copper in sites that have previously showed significant increases (Stephenson and Leonard 1994).

H_0: Additional data from (1990-2000) does not change significant trends in total chlordane, total DDT, and total PCBs levels at the sites.

H_1: Given the more recent data over the past decade (1990-2000), there has continued to be a significant decrease of total chlordane, total DDT, and total PCBs in the sites that have previously showed significant decreases (Stephenson, Martin, and Tjeerdema, 1995).

Objective Two: Determine whether there is a relationship between chemical contaminant concentrations, average rainfall, and land-use (developed, open space, agricultural-row crop, agricultural-no row crop, grasslands, shrub, water, grain, herbaceous wetlands, and mine). Specifically, the null and alternative hypotheses are:

H_0: There is no significant difference between developed land uses and other land use classifications.

H_1: Developed land uses are associated with higher concentrations of heavy metals and organic compounds as compared to other land use classifications.

H_0: There is no significant difference between agricultural land uses and other land use classifications.

H_1: Agricultural land uses are associated with an increase in agricultural chemicals compared to other land use classifications.

METHODS

Overview

Chemical impacts to the marine environment along the California coastline between 1990 and 2000 were analyzed using the California mussel as an indicator for trace metals (copper, lead and silver) and organic compounds (total chlordane, total DDT and total PCBs) contamination. Data for associated trace metal and organic compound concentrations from the sampling locations for resident and transplanted mussels have been provided by SMWP for the years 1977 through 2000.

Site locations used for objective one and objective two in this study are presented in Tables 6 and 7, and in Figure 1. Objective one's 24 sites and objective two's 42 sites are subsets of sites chosen from the 547 sites sampled for metallic and the 472 sites sampled for organic contaminants by the California State Mussel Watch Program (CSMWP). More specifically this subset was drawn from the 20 sites used by M.D. Stephenson and G. H. Leonard in their paper, *Evidence for the Decline of Silver and Lead and the Increase of Copper from 1977 to 1990 in the Coastal Marine Waters of California* (1994) and the 47 sites used by Stephenson *et al.* in their paper *Long-term trends in DDT, Polychlorinated Biphenyls, and Chlordane in California Mussels* (1995).

The sites used for both objectives of this study were divided as follows between resident and transplant mussel sites as set forth in Tables 6 and 7. "Resident sites" are those locations from which California mussels were collected from their native intertidal zone habitat. "Transplant sites" are sites where California mussels, which were previously raised in a clean environment such as Bodega Bay, were relocated to remain at

the site for several months and later collected (Stephenson and Leonard, 1994). The sampling design of the Mussel Watch program is detailed below.

Using Bivalve Mollusks as Indicators

In 1978, Goldberg proposed a nationwide study using mussels as sentinel organisms to monitor chemical contamination and coined the term "Mussel Watch" (Goldberg, 1978; O'Connor, 1998). Mussels are soft-bodied aquatic invertebrates enclosed by two shells connected by a ligament or hinge. They feed by filtering the water that surrounds them, readily accumulating chemical and bacterial pollutants from their surrounding habitat. Due to the mussel's ability to rapidly accumulate both anthropogenic and naturally occurring contaminants, mussels have proven capable of rapidly showing adverse effects of poor water quality. Analyzing mussel tissue for toxic substances has proven to be more accurate than traditional methods of analyzing water samples for chemical concentrations which are often too low to be detected or are temporary in nature and therefore do not provide an accurate picture of the water quality. Mussels are now commonly used throughout the United States to measure the chemical concentrations and impact of wastewater discharges on near shore ecosystems (Haynes, Leeder, and Rayment, 1997).

Many chemicals are not water soluble, but can be accumulated in tissue and sediments. Mussels provide an indication of the average temporal and spatial concentration of bioavailable contaminants in aquatic ecosystems. The levels of

contaminant concentrations can reflect the success of source reduction efforts in a particular watershed (Gunther *et al.*, 1999).

Mussels are valuable long-term indicator organisms. However, there are many biological and environmental factors that affect bioaccumulation and there are important disadvantages to consider (Wilson et al., 1992). For example, mussel bioaccumulation may change with different environmental factors such as food availability, salinity, dissolved oxygen, temperature, and turbidity (Gunther et al., 1999). Sericano, Wade, and Brooks (1996) state that the metabolism of the mussel is too slow to equilibrate with their environment. In the San Francisco Bay, mussels appear to have a limited capacity to accumulate contaminants in the environment, most notably mercury and selenium (Gunther *et al.*, 1999, Luoma and Linville, 1995). These factors must be addressed when analyzing contaminant data bioaccumulated in mussels.

Mussels were chosen as ideal indicators for measuring the level and extent of chemical contamination because they have the following advantages:

- Mussels are sessile and are consequently better indicators of chemical pollution for a specific location than mobile, transient organisms, such as fish (Farrington *et al.*, 1983; O'Connor, 1998, Rasmussen, 1996).
- Mussels respond to metal sources in dissolved and suspended phases (Rainbow, 1995).
- Mussels concentrate chemicals from the surrounding habitat by factors of 10^2 to 10^5 compared to seawater. This ability to bioaccumulate chemicals in their tissues makes measuring for trace constituents simpler than taking water samples (Farrington *et al.*, 1983, Martin and Richardson, 1991; Dame and Allen, 1996; Rasmussen, 1996).
- Mussels are resistant to chemical contamination and survive in polluted waters that generally severely decrease or eradicate other species (O'Connor, 1998, Farrington *et al.*, 1983).
- Mussels provide scientists a valuable evaluation of biological availability of chemicals to organisms (Farrington *et al.*, 1983, Rasmussen, 1996).

- Mussels are found in a variety of diverse geographical areas; thus alleviating problems commonly found in comparing data on considerably different species (Farrington *et al.*, 1983).
- Established local populations of mussels tend to be large enough to be sampled repeatedly, yielding temporal changes in chemical concentrations (Farrington *et al.*, 1983).
- Mussels can be, "successfully transplanted and maintained on sub tidal moorings or on intertidal shore areas where normal populations do not grow-most often due to lack of suitable substrate-thereby allowing expansion of areas to be investigated (Farrington *et al.*, 1983, Rasmussen, 1996)."
- In comparison to other organisms, such as fish, a mussel's enzyme system has a limited ability to metabolize and depurate accumulated contaminants such as aromatic hydrocarbons and PCBs, which make them good indicators of the magnitude of xenobiotic contamination in their surrounding habitat (Phillips, 1997, Farrington *et al.*, 1983, Kim *et al.*, 2001).
- Since mussels are valued worldwide as commercially prized seafood, assessments of chemical contamination are important for public health reasons (Farrington *et al.*, 1983).
- Mussels and macroalgae are currently the most efficient and reliable indicators (Phillips, 1997), providing a relatively inexpensive means to evaluate the relative bioavailability of the metals for each mussel watch site (Phillips, 1997; Munoz-Barbosa, Gutierrez-Galindo, and Flores-Munoz, 2000).

Site locations

The sampling sites in this study reflect a range of land-uses from rural, relatively undisturbed sites to those receiving wastewater directly from sewage treatment plants. The Mussel Watch sampling sites are not uniformly distributed along the coast of California. Sampling locations are generally located along the outer coast, at mouths of estuaries, and within harbors. Sites are on average 20 kilometers (km) apart within estuaries and embayments and approximately 70 km apart along the open coastlines (O'Connor and Beliaeff, 1995, and Lauenstein, Harmon, and Gottholm, 1993). The sampling sites have been chosen primarily by the six coastal RWQCBs to be

representative of their surrounding area (Rasmussen, 1996; O'Connor, 1996). Known point sources of waste discharge and small-scale clusters of contamination have been avoided.

Collection and Preparation of Mussel Tissue for Chemical Analysis

Under a contract with the SWRCB to conduct the fieldwork and laboratory analysis, CDFG personnel collect approximately 100 California Mussels at each sampling site on the California coast. The mussels are usually collected from mussel beds or piers. Transplanted mussels raised in a "clean" collection area, such as Bodega Bay (site 202.0) in Northern California and Montana del Oro (site 430.0) in Central California, are also used in the monitoring program. Transplanted mussels are placed in mesh sacks and secured to stationary objects, such as piers, several feet below low tide for three to five months before being collected for analysis. In order to prevent the transplanted mussels from being washed away from heavy winter rains, the mussels are regularly deployed in late summer and harvested in early winter. The harvested mussels are then randomly divided into two groups to be analyzed for trace metal and synthetic organic chemical analysis (Rasmussen, 1996). Three analytical replicates of 15 mussels are completed and a mean value is established for trace metals on a dry weight basis (parts per million for metals and as parts per billion for organics) (Rasmussen, 1996). A single replicate consisting of 45 mussels is composited for the analysis of synthetic organic compounds (Rasmussen, 1996).

Using the same protocols, three separate laboratories have analyzed the California Mussel Watch data. The University of California's Bodega Marine Laboratory in Bodega, California analyzed the tissue samples from 1977 and 1978. The CDFG Analytical Laboratory in Sacramento, California analyzed the samples from 1979-1986. The tissue was analyzed by the University of California at Santa Cruz and the CDFG Cooperative Trace Organics Analytical Laboratory from 1987-2000 (Stephenson, Martin, and Tjeerdema, 1995).

In order to minimize size-related effects in the analysis, mussels of equal size (55-65 mm in length) are chosen for collection. The mussels are gathered from the highest tidal height that they can be found in adequate numbers in order to diminish variability influenced by habitat height (Martin *et al.*, 1988 and Rasmussen, 1996). The mussels' gonads are dissected and discarded to minimize the differences in body weight due to reproductive cycle seasonality (Martin *et al.*, 1988).

Resident and transplanted mussels are commonly of different species and are known to bioaccumulate at different rates. Resident mussels from a contaminated area will often readily depurate contaminants from their tissue. Therefore, the contaminant levels in transplanted mussels are often higher than the levels in residential mussels. There is concern among researchers as to whether data from the different species of mussels and the different sampling techniques should be combined to address general trends and make comparisons (Birosik, personal communication). Although differences between sites are not known whether to truly exist or be a function of the species bioaccumulation.

OBJECTIVE ONE: IDENTIFYING TEMPORAL CONTAMINANT TRENDS

METHODS

Description of Data Sources

From 1977 through 2000, the SMWP sampled 547 stations for trace metals and 472 stations for organic contaminants. In this study, a subset of 24 collection stations was selected for statistical analysis based on the following criteria: (i) the sites are located along the California coast, excluding San Francisco Bay; (ii) the sites have been previously reported in studies by Stephenson *et al.*, (1995) for organic compounds, or Stephenson and Leonard (1994) for trace metals; and (iii) the sites have been sampled a minimum of four times over the past decade (1990-2000). Site comparisons were limited to those with the same species of mussels. However, either resident or transplanted mussels were deployed at different sites were selected for analysis since mussel species differ in their bioaccumulation characteristics. The 24 sites that fit the above criterion are presented in Table 6.

Statistical Analysis

In order to analyze the data, sites where contaminant values are found to be below the detection limit or are "not detected", ND, were given the value zero. To identify temporal trends in the trace metal and organic concentrations in mussels from 1977 to 2000, regressions (linear, exponential or polynomial to the third power) were performed using SPSS with concentration as the dependent variable and time as the independent variable, for each of the 24 sites. A statistically significant correlation ($p<0.05$) between chemical concentration and time is defined as a trend (O'Connor, 1996). Polynomial

models up to the third power were used to avoid over-fitting. The total slope of the regression was compared between the years 1977–1990 and 1990-2000 for trace metal concentrations in mussels to identify any continuing trends. For organic compound concentrations, the total slope of the regression was compared between the years 1977–1992 and 1992-2000.

RESULTS

Objective One: Long-Term Trends in Trace Metal Concentrations

The hypotheses that given the more recent data over the past decade (1990-2000), there has continued to be a significant reduction of lead and silver in sites that previously showed significant decreases and that there has continued to be a significant increase of copper in sites that have previously showed significant increases (Stephenson and Leonard 1994) were partially substantiated.

Of the 24 sites analyzed, 11 (46%) of the sites showed a clear trend in at least one heavy metal (see Tables 8-10, Figures 2-6). Of the 24 sampling sites, 7 (29%) had significant decreases in silver over time (Table 8). Six of the 24 sampling sites (25%) had significant decreases in lead over time (Table 9). Six of the 24 sampling sites (25%) exhibited increases in copper while only one site showed a decrease in copper (Table 10). Details on the specific elemental trends at the individual sites will be examined below, followed by a discussion of the results. We only considered trends that are significant ($p<0.05$). Sites that did not exhibit temporal trends have not been graphed. Tables 8 through 10 list chemicals for which there is a 95% probability that the slope is significantly different from zero and whether the trend overtime is increasing (I) or decreasing (D).

Silver:

Consistent with Stephenson and Leonard (1994) findings of trace metal trends in 1990, sites 202.0, 414.0, and 430.0 continued to show linear decreases in silver through 2000 (see Table 8, Figures 2-3). As noted by Stephenson and Leonard (1994), site 750 declined over time from 1977 through 1984 followed by a steady increase through 1987. From 1987 until 1989, the concentration of silver dropped again only to be followed by another increase in 1990. From 1990 until 1997, silver concentrations once again rapidly declined. Despite the fact that the last sampling data points increased, the overall trend is a decreasing linear trend.

No significant relationship was found for silver at sites 2.0, 3.0, 616.0, and 715.0. Stephenson and Leonard (1994) also found no significant relationships at sites 3.0 and 616.0, which were consistent with this study's non-significant findings. In contrast to Stephenson and Leonard's non-significant findings for sites 10.0 and 662.0, the additional sampling dates reviewed in this study provided enough data to depict significant linear decreasing trends for these two sites over time. A slight increase was noted in the silver concentration at site 10.0 in 1999, but overall it exhibited a decreasing linear trend. With the removal of the sample data point taken on January 21, 1982, which appeared to be a possible outlier, the data continued to show a decreasing trend, but with a stronger exponential decline. Site 662.0 had very scattered data points, but an over all decreasing linear trend was observed.

Site 894.0 was the only site in the previous study by Stephenson and Leonard (1994) that showed a strong linear increasing trend from 1982 through 1990. Surprisingly, since 1990, this site has rapidly decline to 0.402 ppm (parts per million) of silver, which is lower that the 0.677 ppm exhibited in 1982. With the additional data from the 1990's, this site exhibits a non-linear decrease. Significant values of the regressions ranged from ≤ 0.000 to 0.0329 and the R^2 values ranged from 0.293 to 0.744.

Copper:

In contrast to the majority of the sites showing decreases in silver concentrations, copper concentrations exhibited significant increases over time at 6 of the 24 (25%) sites examined (Sites 2.0, 202.0, 414.0, 430.0, 715.0, and 894.0) (see Table 10, Figures 4 & 5).

Sites 202.0, 414.0, 430.0, 715.0, and 894.0 continued to exhibit increasing trends in copper, which was consistent with Stephenson and Leonard (1994) results.

In contrast to Stephenson and Leonard's non-significant findings for site 2.0, the additional sampling dates collected in this study provided enough data to depict a significant linear increasing trend over time. Stephenson and Leonard (1994) found site 3.0 to have no significant trend overtime; however, with the additional data, the site showed a non-linear decreasing trend. Site 3.0 was the only site that exhibited a non-linear decrease in copper overtime. Interestingly, from 1989 through 1997 the site showed substantial increases in copper.

No significant relationship was found for copper at sites 10.0, 616.0, 662.0, and 750.0, which was consistent with Stephenson and Leonard's non-significant findings.

Significant values of the regressions ranged from 0.0151 to 0.0537 and the R^2 values ranged from 0.143 to 0.601.

Lead:

Unlike copper, lead steadily decreased over time at six of the 24 sites (25%). Sites 202.0, 616.0, 662.0 and 750.0 continued to show decreases in lead through 2000, which was consistent with Stephenson and Leonard's decreasing findings (see Table 9, Figure 6). In comparison to Stephenson and Leonard's non-significant findings for sites 414.0 and 715.0, the additional sampling dates reviewed in this study provided enough data to depict linear decreasing trends for both sites. However, with the additional sampling data, sites 430.0, 721.0, 723.0, and 726.0 were no longer showing significant linear decreases as they previously did in Stephenson and Leonard's study. They all had no significant trends. No significant relationships were found for sites 2.0, 3.0, 10.0, and 894.0. Stephenson and Leonard (1994) also did not find any significant relationships at sites 2.0, 3.0, 10.0 and 894.0, but noted a decreasing trend at site 430.0. It is important to mention that no sites showed increasing trends over time. Significant values of the regressions ranged from ≤ 0.000 to 0.0278 and the R^2 values ranged from 0.147 to 0.835.

Objective One: Long-Term Trends in Organic Contaminants

The hypotheses that given the more recent data over the past decade (1990-2000), there has continued to be a significant decrease of total chlordane, total DDT, and total

PCBs in the sites that have previously showed significant decreases (Stephenson, Martin, and Tjeerdema, 1995) were partially substantiated.

Nineteen of the 24 sites (79%) showed a clear trend in at least one organic contaminant (see Tables 11-13). Five of the 24 sites (21%) exhibited no relationship for any organic contaminant. Of the 24 sampling sites, twelve (50%) had significant decreases in total chlordane over time, which was consistent with Stephenson, Martin, and Tjeerdema's (1995) findings of 14 of 28 sites (50%) showing decreases in total chlordane (see Table 11, Figures 7, 8, & 9). Twelve of the 24 sites (50%) exhibited significant decreases in total DDT, compared to Stephenson, Martin, and Tjeerdema's (1995) findings of 47% (15 of 32) displaying decreases in total DDT (see Table 12, Figures 10, 11 & 12). Only one of this study's 24 sites (4%) showed significant increases in total DDT whereas Stephenson, Martin, and Tjeerdema (1995) did not report any increasing trends for total DDT. Ten of the 24 sites (42%) exhibited significant decreases in total PCBs (see Table 13, Figures 13, 14, 15, & 16). Seven of the 24 sites (29%) exhibited significant increases in total PCBs, which were in complete contrast to Stephenson, Martin, and Tjeerdema's (1995) report where they found no significant increases in total PCBs.

Details on the specific elemental tends at the individual sites will be examined below followed by a discussion of what the results could imply. Sites that did not exhibit significant temporal trends have not been graphed.

Total chlordane:

Of the 24 sites examined, nine sites were not analyzed for total chlordane because they did not meet the study's criteria for statistical analysis. Of the remaining 15 sites, 12 (Sites 10.0, 202.0, 414.0, 601.0, 616.0, 662.0, 713.0, 715.0, 724.0, 725.0, 726.4, and 750.0) had statistically significant decreases in total chlordane over time (see Table 11, Figures 7, 8, & 9). Sites 202.0, 414.0, 601.0, 616.0, 662.0, 713.0, 715.0, 724.0, 725.0, and 750.0 continued to show decreases in total chlordane through 2000, which was consistent with Stephenson, Martin, and Tjeerdema's (1995) decreasing findings for these sites. Site 404.0 was an exception. This site previously exhibited a decreasing trend in the 1980's; however, with the additional data an increasing trend was observed in the 1990's. Stephenson, Martin, and Tjeerdema (1995) observed no significant relationships for sites 10.0, 601.0, 616.0, 713.0, 715.0, and 726.4, but with the additional data from the 1990's these sites exhibited decreasing trends.

Sites 2.0, 3.0, 100.0, 104.5, 744.6, and 894.0 showed no significant relationship for total chlordane. Stephenson, Martin, and Tjeerdema (1995) also did not find any significant relationships at sites 2.0, 3.0, 104.5, 744.6, and 894.0. Interestingly, only two sites (103.0 and 404.0) showed non-linear increasing trends in the 1990's. Significant values of the regressions ranged from ≤ 0.000 to 0.023 and the R^2 values ranged from 0.411 to 0.913.

Total DDTs:

Of the 24 sites examined, seven sites were not analyzed for total DDT because they did not meet the criteria for statistical analysis. Of the remaining 17 sites, 12 (50 %) (sites 10.0, 100.0, 103.0, 601.0, 616.0, 662.0, 713.0, 715.0, 724.0, 725.0, 726.4 and 750.0) had statistically significant decreases in total DDT over time (see Table 12, Figures 10, 11 & 12). Sites 10.0, 601.0, 616.0, 713.0, 715.0, 724.0, and 725.0 continued to show decreases in total DDT through 2000, which was consistent with Stephenson, Martin, and Tjeerdema's (1995) decreasing findings for these sites. Sites 100.0, 662.0, and 726.4 exhibited decreasing trends, where in comparison Stephenson, Martin, and Tjeerdema (1995) found site 100.0 to have more than 80% of the samples below the detection limit and found no significant relationships for sites 662.0 and 726.4.

This study found no significant relationship for total DDT at sites 2.0, 3.0, 104.5, 202.0, 336.0, 414.0, 744.6, and 894.0. Stephenson, Martin, and Tjeerdema (1995) also did not find any significant relationships at sites 2.0, 3.0, 202.0, 336.0, 414.0, 662.0, 724.6, 744.6, 750.0, and 894.0. It is noteworthy that site 404.0.0 is the only site that showed non-linear increases in the 1990's. This site 404.0 previously exhibited decreases in the 1980's (Stephenson, Martin, and Tjeerdema, 1995). Significant values of the regressions ranged from ≤ 0.001 to 0.058 and the R^2 values ranged from 0.209 to 0.825.

Total PCBs:

Of the 24 sites examined, one site (430.0) was below the detection limit. Of the remaining 22 sites, 7 (29%) (sites 10.0, 202.0, 404.0, 414.0, 443.0, 744.6, and 894.0) had statistically significant increases in total PCBs over time (see Table 13, Figures 13, 14, 15, & 16). Interestingly, sites 10.0, 202.0, 414.0, and 443.0 had many sampling dates in the mid-eighties to early nineties where the data concentrations were below the detection limit. In plotting these data points, they all shared similar parabola-shaped trend lines where the concentration of total PCBs started high, decreased down to below the detection limit and then increased again in the 1990's.

With the additional sampling data from the 1990's, sites 10.0, 202.0, 443.0, and 744.6 were all found to have increasing trends, where previously Stephenson, Martin, and Tjeerdema (1995) found these sites to have more than 80% of the samples below the detection limit.

In contrast, sites 103.0, 444.0, 601.0, 616.0, 662.0, 713.0, 715.0, 724.0, 725.0, and 750.0 exhibited significant decreases in total PCBs over time. Sites 662.0, 725.0, and 750.0 continued to show decreases in total PCBs through 2000, which was consistent with Stephenson, Martin, and Tjeerdema's (1995) decreasing findings for these sites. Stephenson, Martin, and Tjeerdema (1995) found sites 103.0 and 444.0 to have more than 80% of the samples below the detection limit. With the additional data from the 1990's, site 103.0 exhibited a significant non-linear decrease overtime and site 444.0 exhibited a significant linear decrease. No significant relationships were previously found by Stephenson, Martin, and Tjeerdema (1995) at sites 616.0, 715.0, and 724.0; however,

with the additional data from the 1990's, these sites showed significant linear decreases overtime.

The nine remaining sites (2.0, 3.0, 100.0, 104.5, 336.0, 434.0, 435.0, 437.0, and 726.4) exhibited no significant relationship for total PCBs over time. Significant values of the regressions ranged from ≤ 0.000 to 0.026 and the R^2 values ranged from 0.252 to 0.855.

DISCUSSION

NOAA Researcher Thomas P. O'Connor (1996) stated that a trend is the single attribute of the Mussel Watch data that distinguishes natural from anthropogenic influences. Trends identified in his 8-year study could have natural causes only if there has been a systematic shift in salinity, reproduction, growth rate or a combination of such variables for over 6 years forcing a parallel shift in chemical concentrations in mussel tissues (O'Connor, 1996). As O'Connor (1996) noted, a trend due to a unique coincidence of natural factors seems even less probable.

Silver:

The decrease in silver in mussels is probably due to a combination of factors. In previous studies, researchers have suggested that the decreasing trends in silver were most likely related to the presence and increased reclamation efforts of wastewater treatment plants (Stephenson and Leonard, 1994; and O'Conner, 1992). Waste management facilities have realized the monetary value in reclaiming silver from discharge water. Stephenson and Leonard (1994) believe that decreased mass emission rates by wastewater treatment plants appear related to the decline in silver concentrations in mussels. Silver input to the ocean from outfalls decreased 62% from 1984 to 1987, which may have played a role in the decrease in silver concentrations in mussels.

Site 414.0 (Pacific Grove) is located near an inactive wastewater treatment plant (WWTP) outfall. The WWTP closed in 1980 and silver concentrations in mussels have

continued to decrease since then (Stephenson and Leonard, 1994). Site 662 (Royal Palms) showed significant linear decreases in silver overtime which may reflect a decrease in silver mass emissions from the large municipal outfall that is in close proximity to the mussel site.

Sites 202 (Bodega Bay) and 430 (Montana De Oro) exhibited significant linear decreases in silver overtime. No known sources of silver are present at either of these two sites. Historically, silver has been very low at these sites (Stephenson and Leonard, 1994).

Site 894 (San Diego Bay) was the only site to show a non-linear decrease in silver. Stephenson and Leonard previously found this site to be increasing significantly throughout the 1980's. They believed that runoff from San Diego airport and nearby aeronautical industries may have contributed to this increase in silver. Better reclamation efforts by these industries may have caused a reduction of silver runoff to this mussel site.

The declines in silver could also be attributed to the fact that photography industries are operating under standards that are more stringent and are improving their reclamation efforts. With the increase use of digital cameras, we will begin to see the demand for silver decrease (Craig, Vaughan, and Skinner, 1996) and the further reduction of silver waste from photo development.

Copper:

Copper makes up a small percent of the Earth's crust (0.0058 percent) and is emitted naturally into air and water through wind blown dust, volcanic eruptions, and the weathering of soil (Lomborg, 2001; ATSDR, 1990). Copper may also enter the air through anthropogenic activities such as copper smelting and ore processing, fungicide use on plants or in waterbodies to control algae, and application to wood, fabric, and leather. Copper also enters into waterways through industry discharges, sewage treatment plants emissions, and copper-based boat paints (ATSDR, 1990). Another possible source of copper is automobile brake pads. With the increase in population, there are more cars on the road, and it is likely that urban runoff is bringing contaminants, such as copper dust from the wearing of brake pads into our waterways and out to the ocean where aquatic organisms can absorb them.

It is very interesting that six of the seven significant trends in copper concentrations were increasing trends. This study's findings were consistent with the increases found in the report by Stephenson and Leonard (1994). Stephenson and Leonard (1994) and Lauenstein, Robertson, and O'Connor, 1990) believe the increase in copper concentrations in mussel tissue is likely due to the increased usage of copper since the 1970's.

No known sources of copper exist at sites 414 (Pacific Grove), 715 (Huntington Harbor) and 894 (San Diego Bay). However, Stephenson and Leonard (1994) believed that the increase in copper levels could be a result of the increase in boat traffic and the use of anti-fouling paints containing copper to help prevent organisms from attaching to

the hulls of boats. These new copolymer paints consist up to 30% copper. The increase in copper at sites 715 (Huntington Harbor) and 894 (San Diego Bay) could be due to the increased vessel traffic and the releasing of copper in the water column and absorption in mussel tissue. Site 414.0 (Pacific Grove) is in the Monterey Bay and could be influenced by the boating activity in and around Monterey Harbor.

Site 2.0 (Crescent City) exhibited decreases in copper concentrations from 1983 through 1989, followed by increases in copper from 1989 through 1997 and it is unclear why such increases were observed. One could speculate that the increases are linked to increases in copper effluent from the outfall at Crescent City, but further investigations would need to be done.

It is also unclear as to why sites 202.0 (Bodega Head) and 430.0 (Montana de Oro) are experiencing increasing trends in copper concentrations. Site 202.0 shows significant decreases in silver and lead as well, which raises questions as to what is happening at this particular site. Site 3 (Crescent City) was the only site to show a decreasing trend and the reason is uncertain as to why it would be decreasing.

Lead:

Declines in lead most likely are a result of the phase-out of lead additives in gasoline, decreased mass emission rates by WWTPs, and improved reclamation efforts (Stephenson and Leonard, 1994; Squire *et al.*, 2002). Lead-based paints were banned in 1978, followed by the banning of leaded gasoline in 1992, which is reflected in the decreasing lead concentrations in the tissues of mussels throughout the 1980's and

1990's. In addition, the public awareness campaigns regarding the health risks associated with lead, specifically neurotoxicity in children, the regulatory restrictions on the use and manufacture of lead have helped reduce the usage of lead in the United States.

Site 202.0 (Bodega Head) showed significant declines in lead, but again it is unclear as to what is influencing such declines in lead, silver and copper concentrations. Site 414.0 (Pacific Grove) exhibits a flatter linear decrease in lead concentration overtime, which may be indicative of the lower levels of runoff containing lead.

Sites 616.0 (LA Harbor), 662.0 (Royal Palms), and 715.0 (Huntington Harbor) are bordering highly developed, populated areas and are perhaps being influenced by local runoff that is elevating the levels of lead in the water.

Total chlordane:

With two exceptions, sites 103.0 (Eureka Channel) and 404.0 (Sandholdt Bridge), total chlordane was showing decreasing trends overtime. These decreases in total chlordane are expected and are likely due to the fact that total chlordane was banned in 1988. Stephenson, Martin and Tjeerdema, (1995) stated that the declining levels of chlorinated organics, such as chlordane, in the tissue of mussels, are reflective of the regulatory restrictions on their use and production.

They also suggested that harbors might be experiencing decreases in organic contaminants such as chlordane because contaminated sediment is being covered by cleaner, more recent sediment. This could be the case with sites 601.0 and 616.0 in LA

Harbor, 713.0 and 715.0 in Huntington Harbor, and sites 724.0, 725.0, and 726.4 in Newport Bay Harbor, all of which exhibited declining levels of chlordane overtime.

Stephenson, Martin and Tjeerdema, (1995) noted that the dredging of harbors also removes contaminated sediment. However, site 404.0 (Sandholdt Bridge) in Moss Landing Harbor, surrounded by agricultural communities, showed no declines of chlordane. In fact, interestingly, levels of chlordane increased over time, indicating that the chlordane is being re-suspended in the environment. Most likely, the rapid expansion of neighboring communities, such as Salinas and Watsonville, have caused the soil to be disturbed and has lead to increased urban and agricultural runoff.

Total DDT:

Like chlordane, declining levels of DDT is expected and probably due to the banning of the domestic use of DDTs in the United States back in 1972. All significant trends were exhibiting decreases overtime, except one site, site 404.0 (Sandholdt Bridge) in Moss Landing, which exhibited an increasing trend overtime in DDT. It is recommended that this site be investigated closely to determine what areas have more contamination. Sediment samples in nearby neighborhoods would be very valuable and may shed some light as to where DDT is coming from. Most likely, the development in the surrounding communities is remobilizing agricultural soil containing DDT. Several harbor sites showed significant linear decreases in DDT, which once again may be a result of contaminated DDT sediment being covered by cleaner, more recent sediment.

Total PCBs:

Ten sites (103.0 Eureka Channel, 444.0 Intake Cove, 601.0 LA Harbor, 616.0 LA Harbor, 662.0 Royal Palms, 713.0 Huntington Harbor, 715.0 Huntington Harbor, 724.0 Newport Bay Harbor, 725.0 Newport Bay Harbor, and 750.0 Oceanside) exhibited significant decreases in total PCBs over time, which are reflective of the fact that total PCBs were banned in 1976. However, seven sites (10.0 Trinidad Head, 202.0 Bodega Head, 404.0 Sandholdt Bridge, 414.0 Pacific Grove, 443.0 Diablo Cove, 744.6 San Onofre 6, and 894.0 San Diego Bay) had statistically significant increases in total PCBs over time, which raises questions as to the source of PCBs. Again, it is possible that the excavation and development in the surrounding communities is remobilizing agricultural soil and bottom sediments containing PCBs. The weathering of old asphalt, the leakage of old electronic equipment, and the leaching of landfills may also be contributing sources of PCBs. PCBs degrade very slowly and are consequently still persistent in the environment.

It is unclear as to why sites 10.0 (Trinidad Head), 202.0 (Bodega Head), 414.0 (Pacific Grove), and 443.0 (Diablo Cove) had many sampling dates in the mid-eighties to early nineties where the data concentrations were below the detection limit and then increased again in the 1990's. Further investigation is needed.

OBJECTIVE 2: DETERMINING THE EFFECTS OF LANDUSE AND RAINFALL ON SILVER, COPPER, LEAD, TOTAL CHLORDANE, TOTAL DDTS, AND TOTAL PCBS

METHODS

Description of Data Sources

For Objective 2, a subset of 42 collection stations was selected for statistical analysis based on the following criteria: (i) the sites are located along the California coast. (ii) The sites have been previously reported in studies by Stephenson, Martin and Tjeerdema, (1995) for organic compounds, or Stephenson and Leonard (1994) for trace metals. (iii) The sites have been sampled a minimum of four times over the past decade (1990-2000) for both trace metals and organics. Site comparisons were limited to those with the same species of mussels, since mussel species differ in their bioaccumulation characteristics. The 42 sites that fit the above criterion are presented in Table 7.

Contaminant data was obtained from the SMWP (see Table 14).

Historical precipitation data were obtained from the Western Regional Climate Center (WRCC) in Reno, Nevada. The precipitation values for the four months prior to sampling in addition to the month in which the sample was taken were used to conduct the analyses, since the California mussels respond rapidly to changes in pollutant body burden.

Watershed maps provided by the Department of Water Resources were compiled and put into a Geographic Information System (GIS) in order to identify the various watersheds surrounding the Mussel Watch sites (see Figure 17). Land-use data produced

by the U.S. Geological Survey (USGS) and the EPA provided the land cover data layer used in the GIS as well as in the multiple regression analysis. This National Land Cover Characterization Project was based on 30-meter Landsat Thematic Mapper ™ data.

Statistical Analysis

In order to estimate the impact of land use and rainfall on the contaminant concentration observed in mussels, a stepwise multivariate regression statistical technique was used with concentration as the dependent variable and land use and rainfall as the independent variables (Table 15).

Contaminant data was obtained from the SMWP. The arithmetic mean was calculated for the 42 sites.

The original land cover data set from USGS and the USEPA was classified into 21 different land covers/land-uses for the state of California, encompassing low intensity residential to high intensity residential land cover to a variety of forested and cultivated classifications. Since this study focused on the coastal, temperate watersheds, the land-use category for "Ice and Snow" was disregarded for this analysis. The remaining 20 independent (predictor) variables were then auto-correlated to detect any differentiation between each land-use. To help reduce the number of land-uses, a Principal Components Factor Analysis with Varimax Rotation was applied (using SPSS). Any missing values were replaced with the mean m^2 of the particular land-use. Based on the statistical results and on the knowledge of similarities between land-uses, all variables that were found to co-vary together were grouped together under one of seven types of land-use categories:

developed, open space, agricultural-row crop, agricultural-no row crop, grasslands, shrub, and water. Those land-use categories that were not found to co-vary with another land-use were assigned into one of the seven designations or eliminated from the analysis. The land-use categories "Bare Rock, sand, clay", "Quarries, Strip Mines and Gravel Pits" and "Emergent Herbaceous Wetlands" were eliminated. "Rock, sand, clay" was discarded because it co-varied with the developed land-use categories. "Quarries, Strip Mines and Gravel Pits" and "Emergent Herbaceous Wetlands" were eliminated because they did not co-vary with the other groups and the author specified seven land-use groups. These seven land-use groups that were used as independent variables are described in Table 16.

Using GIS, the specific watersheds surrounding each mussel watch site were determined and the total square meters of each 19 land covers were compiled into a large database, aggregated and used in the stepwise multivariate regression as independent variables. The goal was to see what different land covers were present in each watershed and potentially influencing the chemical concentrations in the nearby mussels.

The rainfall data for the four months prior to sampling in addition to the month in which the mussel was collected was gathered for each sampling date in order to calculate the average and total amount of rainfall for each sampling date for all of the 42 sites (see Table 17). The various averages were calculated and then auto-correlated to detect any differentiation between each rainfall average. All rainfall variables were found not to be significantly correlated with each other, so the average rainfall for each site was chosen as the independent variable for statistical analysis.

RESULTS

Objective Two: Correlations between Land-use, Rainfall and Chemical Concentrations

The land-use category, "developed", positively correlated to higher lead (Table 1), total chlordane (Table 2), and total DDT (Table 3) concentrations substantiating the hypothesis that developed land uses are associated with higher concentrations of heavy metals and organic compounds as compared to other land use classifications. The land-use category "LN row crop" positively correlated to higher copper (Table 4) and total chlordane (Table 2) concentrations substantiating the hypothesis that agricultural land uses are associated with an increase in agricultural chemicals compared to other land use classifications.

Lead:

Table 1. Stepwise Regression Coefficients for Lead (ppm) using seven land-uses, average rainfall, and resident and transplanted mussels.

Model Summary

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
1	.517(a)	.267	.249	3.63356
2	.593(b)	.352	.319	3.46019
3	.694(c)	.482	.441	3.13444

a Predictors: (Constant), DEV_1

b Predictors: (Constant), DEV_1, WATER_7

c Predictors: (Constant), DEV_1, WATER_7, OPENSP_2

ANOVA(d)

Model		Sum of Squares	df	Mean Square	F	Sig.
1	Regression	192.202	1	192.202	14.558	.000(a)
	Residual	528.111	40	13.203		
	Total	720.312	41			
2	Regression	253.368	2	126.684	10.581	.000(b)
	Residual	466.944	39	11.973		
	Total	720.312	41			
3	Regression	346.972	3	115.657	11.772	.000(c)
	Residual	373.340	38	9.825		
	Total	720.312	41			

a Predictors: (Constant), DEV_1

b Predictors: (Constant), DEV_1, WATER_7

c Predictors: (Constant), DEV_1, WATER_7, OPENSP_2

d Dependent Variable: PB

Coefficients(a)

Model		Unstandardized Coefficients		Standardized Coefficients	t	Sig.
		B	Std. Error	Beta		
1	(Constant)	2.212	.696		3.177	.003
	DEV_1	7.482E-09	.000	.517	3.815	.000
2	(Constant)	1.333	.769		1.734	.091
	DEV_1	7.197E-09	.000	.497	3.845	.000
	WATER_7	5.863E-08	.000	.292	2.260	.029
3	(Constant)	2.572	.804		3.200	.003
	DEV_1	5.343E-09	.000	.369	2.971	.005
	WATER_7	1.161E-07	.000	.578	3.873	.000
	OPENSP_2	-2.061E-08	.000	-.473	-3.087	.004

a Dependent Variable: PB

As shown in Model three of Table 1, the stepwise regression found development and open water as positively related to lead concentration and open space as negatively related to lead concentration (R-square=0.482, $F(3,38)=11.772$, $p < 0.005$).

Total chlordane:

Table 2. Stepwise Regression Coefficients for Total chlordane (ppb) using seven land-uses, average rainfall, and resident and transplanted mussels.

Model Summary

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
1	.753(a)	.567	.556	22.97843
2	.783(b)	.613	.593	22.01350

a Predictors: (Constant), DEV_1

b Predictors: (Constant), DEV_1, LNROWCRO

ANOVA(c)

Model		Sum of Squares	df	Mean Square	F	Sig.
1	Regression	27665.300	1	27665.300	52.396	.000(a)
	Residual	21120.325	40	528.008		
	Total	48785.625	41			
2	Regression	29886.452	2	14943.226	30.837	.000(b)
	Residual	18899.173	39	484.594		
	Total	48785.625	41			

a Predictors: (Constant), DEV_1

b Predictors: (Constant), DEV_1, LNROWCRO

c Dependent Variable: CHRLDANE

Coefficients(a)

Model		Unstandardized Coefficients		Standardized Coefficients	t	Sig.
		B	Std. Error	Beta		
1	(Constant)	10.241	4.403		2.326	.025
	DEV_1	8.976E-08	.000	.753	7.238	.000
2	(Constant)	-14.801	12.435		-1.190	.241
	DEV_1	7.902E-08	.000	.663	6.127	.000
	LNROWCRO	2.128	.994	.232	2.141	.039

a Dependent Variable: CHRLDANE

As shown in Model two of Table 2, the stepwise regression found development and LN row crop as positively related to total chlordane concentration (R-square=0.613, $F(2,39)=30.837$, $p < 0.005$).

Total DDTs:

Table 3. Stepwise Regression Coefficients for Total DDT (ppb) using seven land-uses, average rainfall, and resident and transplanted mussels.

Model Summary

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
1	.407(a)	.166	.145	405.90870

a Predictors: (Constant), DEV_1

ANOVA(b)

Model		Sum of Squares	df	Mean Square	F	Sig.
1	Regression	1311816.182	1	1311816.182	7.962	.007(a)
	Residual	6590474.750	40	164761.869		
	Total	7902290.933	41			

a Predictors: (Constant), DEV_1

b Dependent Variable: DDT

Coefficients(a)

Model		Unstandardized Coefficients		Standardized Coefficients	t	Sig.
		B	Std. Error	Beta		
1	(Constant)	110.171	77.785		1.416	.164
	DEV_1	6.181E-07	.000	.407	2.822	.007

a Dependent Variable: DDT

As shown in Model one of Table 3, the stepwise regression found development positively related to total DDT concentration (R-square=0.166, $F(1,40)=7.962$, $p<0.05$).

Copper:

Table 4. Stepwise Regression Coefficients for Copper (ppm) using seven land-uses, average rainfall, resident and transplanted mussels.

Model Summary

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
1	.360(a)	.130	.108	21.27228

a Predictors: (Constant), LNROWCRO

ANOVA(b)

Model		Sum of Squares	df	Mean Square	F	Sig.
1	Regression	2695.268	1	2695.268	5.956	.019(a)
	Residual	18100.402	40	452.510		
	Total	20795.669	41			

a Predictors: (Constant), LNROWCRO

b Dependent Variable: CU

Coefficients(a)

Model		Unstandardized Coefficients		Standardized Coefficients	t	Sig.
		B	Std. Error	Beta		
1	(Constant)	-8.559	11.818		-.724	.473
	LNROWCRO	2.159	.885	.360	2.441	.019

a Dependent Variable: CU

As shown in Table 4, the stepwise regression found LN row crop as positively related to copper concentration (R-square=0.130, $F(1,40)=5.956$, $p < 0.05$).

Silver:

Table 5. Stepwise Regression Coefficients for Silver (ppm) using seven land-uses, average rainfall, resident and transplanted mussels.

Model Summary

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
1	.306(a)	.094	.071	.82030

a Predictors: (Constant), R1T2_1

ANOVA(b)

Model		Sum of Squares	df	Mean Square	F	Sig.
1	Regression	2.783	1	2.783	4.136	.049(a)
	Residual	26.915	40	.673		
	Total	29.699	41			

a Predictors: (Constant), R1T2_1

b Dependent Variable: AG

Coefficients(a)

Model		Unstandardized Coefficients		Standardized Coefficients	t	Sig.
		B	Std. Error	Beta		
1	(Constant)	1.358	.452		3.004	.005
	R1T2_1	-.537	.264	-.306	-2.034	.049

a Dependent Variable: AG

As shown in Table 5, the stepwise regression found the resident/transplant variable negatively related to silver concentration (R-square=0.094, $F(1,40)=4.136$, $p < 0.005$). The standardized coefficient value of -0.306 indicates lower concentrations of copper are found in transplanted mussels.

Total PCBs:

The stepwise regressions for total PCBs showed no relationships between the chemical concentrations, the seven land-use variables, average rainfall, and the resident and transplanted mussels.

DISCUSSION

Rainfall:

In this study results indicated that precipitation did not significantly influence the trace metal trends or the organic compound trends. The methods used could have too much variability that make it difficult to detect rainfall effects. Perhaps this is in part due to the fact that the rainfall data was not representative of first rainfalls or storm events when the contaminate load on watersheds can be higher. Harte *et al.* (1985) stated that water quality problems linked to urban runoff almost certainly appear following the first fall rains, "flushing land surfaces of metals that have accumulated over the dry Californian summers." The rain data were collected for the four to five months prior to mussel collection, which ranged from early fall to late winter or spring of each year thereby missing the potential influence of first rainfalls.

The land-use category "developed" positively correlated to higher lead (Table 1), total chlordane (Table 2), and total DDT (Table 3) concentrations partly supporting the hypothesis that developed land-uses are associated with higher concentrations of heavy metals as compared to other land-use classifications. Total PCBs, silver and copper showed no correlations with developed land-uses.

The land-use category "LN row crop" positively correlated to higher copper (Table 4) and total chlordane (Table 2) concentrations partly supporting the hypothesis that agricultural land-uses are associated with an increase in agricultural chemicals compared to other land-use classifications. Total DDT showed no correlation with agricultural land-uses.

Silver:

As stated earlier in this paper, resident and transplanted mussels are commonly of different species and are believed to bioaccumulate chemicals at different rates. Sericano *et al.* (1990) found that resident mussels from a contaminated area will often readily depurate contaminants from their tissue and as a result the contaminant levels in transplanted mussels are often higher than the levels in residential mussels. In contrast to Sericano *et al.*'s (1990) findings, my study found that transplanted mussels exhibited lower levels of silver than residential mussels. Silver was the only chemical that exhibited a significant correlation with residential and transplanted mussels (Table 5).

This correlation may be because residential mussels are located at more contaminated sites or that they are capable of accumulating more contaminants than transplanted mussels given the same environmental conditions. Another explanation could be that residential mussels have been stationary at the particular sampling site and consequently have been in the local water for a longer period than the transplanted mussels, which originate from other locations. Further investigation is needed to address this dilemma. A controlled experiment where both residential and transplanted mussels were deployed at the same sampling location would help solve the questions raised.

Copper:

As seen in Table 4, for every $1m^2$ increase in LN row crop land-use, copper increases by 2.16 ppm. This is an interesting result, which leads one to speculate that fungicidal use on plants could be a significant source of copper. In particular, copper

sulfate (*pentahydrate*) is commonly used as a germicide and fungicide. It is extensively used on rice crops to control tadpole shrimp (*Triops longicaudatus*). In fact, the Department of Pesticide Regulation (DPR, 1998) reported that nearly all copper sulfate (90 percent in 1995) was used on rice. Since rice fields are commonly located in California's central valley, and not along the California coast, it leads one to believe that copper sulfate among other copper-based pesticides are being used on other types of row crops. The total use of copper on agricultural fields merit further study.

Lead:

As seen in Table 1, lead has the following significant correlations: 1) for every $1m^2$ increase in developed land-use, lead increases by 5.34×10^{-9} ppm; 2) for every $1m^2$ increase in open water land-use, lead increases by 1.16×10^{-7} ppm; and 3) for every $1m^2$ increase in open space land-use, lead decreases by 2.06×10^{-8} ppm. These correlations imply that lead is a ubiquitous contaminant in the environment, especially in developed areas where lead has been historically used in household paints, plumbing materials, and in gasoline. The correlation between lead and areas of open water, such as lakes and estuaries, may imply that urban runoff carrying trace amounts of lead from industrial waste sites and residential areas is contaminating the waterways and consequently the tissue of coastal California mussels. The negative correlation of lead with open space is quite understandable. One would not expect as much lead being present in open space areas, such as rural grasslands, as in an urban setting.

Total chlordane:

As seen in Table 2, for every $1m^2$ increase in developed land-use, total chlordane increases by 7.9×10^{-8} ppb. For every $1m^2$ increase in LN row crop, total chlordane increases by 2.13 ppb. Assuming LN row crop is kept constant, total chlordane increases by 0.663 standard deviations for every standard deviation increase in developed square meters. The correlations between total chlordane and developed and agricultural areas may represent the heavy historical use of total chlordane as a pesticide on agricultural crops and residential gardens. Since 1948, Total chlordane was extensively used as a pesticide and a fumigant for controlling termites on residential homes and office buildings until it was banned in 1988. Since total chlordane can remain in the soil for over 20 years, it is probably being re-suspended into the environment through processes such as erosion and land excavating.

Total DDT:

As seen in Table 3, the results show that for every $1m^2$ increase in developed land-use, total DDT increases by 6.18×10^{-7} ppb. Like chlordane, the correlation between DDT and development might be due to the fact that many watersheds are undergoing a conversion from pastoral/agricultural land-uses to suburban land-uses (Keller, interview on October 16, 2001). DDT is a well-recognized toxic, manmade insecticide, which dates back to 1938, when it was first introduced as a miraculous insecticide. Before being banned in 1972, it was extensively used worldwide to control against malaria, typhus and other insect carried diseases. In the U.S. alone, approximately 675,000 tons of DDT was

used for intensive agricultural and commercial treatment (Harrison, 1997). With the heavy application of DDT everywhere, there is no doubt that DDT is ever-present in the environment and is probably being re-suspended with any form of land disturbance.

Total PCBs:

The stepwise regressions showed no relationships between chemical concentrations and land-use variables.

Overall Limitations:

As with all research, the limitations of this design are many. The statistical design for this study uses the aggregation method for determining the averages of the concentration, land-use and rainfall data on an individual basis. Averaging the concentration data artificially reduces the variability. This is a problem with analyzing data that is of great variability in nature.

There are numerous environmental factors that influence the California mussel, such as food availability, salinity, dissolved oxygen, temperature, and turbidity, which are difficult to measure and evaluate and which may affect contaminant concentrations. There are limitations in the interpretation of results due to biological variability of mussels, such as genetic polymorphism seen in natural populations and the complexity in determining homogenous age classes (Frazier, 1975). Transplanted mussels that have been raised under identical conditions have been used at some stations to minimize biological variability. However, in this study there are no control groups.

There are numerous factors that affect the molluscan concentrations of chemicals. The physiological cycles of the mussels are subject to shifts within one year due to changes in environmental conditions and in particular temperature (Beliaeff *et al.*, 1997). Laboratory experiments in which conditions of exposure are controlled (Cunningham and Tripp, 1975; Fisher and Teyssie, 1986; Pruell *et al.*, 1987) and field experiments in which the mussels are transplanted from sites of low chemical concentrations to sites of high chemical concentration and vice versa (O'Connor, 1996; Rosesijjadi *et al.*, 1984; Martin, 1985; Capuzzo *et al.*, 1989; and Sericano, 1993) have provided evidence that the chemical concentrations in mussels depend upon the chemical concentrations found in the water and food taken in by the mussel.

The Mussel Watch Program attempts to limit variation by collecting the mussels at the same site, within the limited size range, and in the winter before the spawning season. By selecting mussels within a narrow size range of 55-65 mm in length, the initial variability within and among sampling stations is minimized. To reduce variability attributable to water and facilitate comparisons with other mussel studies, the dry weight data was used.

It is noteworthy to consider that due to the Mussel Watch Program's sampling schedule, some of the samplings may not reflect the maximum exposure to chemical contaminants, such as certain pesticides used only during specific growing periods throughout the year. It is also important to note that due to budget constraints the sampling sites are usually sampled no more than once a year and consequently the maximum contaminant concentrations may not reflect missed periods of intense rainfall.

Because these variables are impossible to control, there will be some noise in the data. Noise in the concentrations will be reduced and a more accurate temporal picture of the dynamics of contaminant body burden will develop with subsequent sampling and analyses. Sequential, consistent sampling on a quarterly basis would provide a more accurate temporal picture of the dynamics of contaminant body burden and synergistic toxicological effects. As more samples are taken in the future, the detection of temporal trends will improve and safe environmental concentrations will be able to be identified.

CONCLUSIONS

This thesis leads to several conclusions. First, as is well evidenced in current research, including this thesis, that environmental monitoring as it is currently carried out does allow for the detection of trends of major contaminants. Specifically, such detection is useful in identifying spatial and temporal patterns of contamination along the California coast.

Second, many of the organic and non-organic contaminants entering our environment may adversely affect human health. Through history the continual increase in population and agricultural and industrial development have negatively impacted the environment. This trend continues today. By monitoring the amount of contaminants entering the environment, we can determine if an area is contaminated with certain potentially toxic substances, identify the sources of such contamination, warn the public of highly contaminated areas, and identify areas that may need to be 'cleaned up'. The more uncontrolled variables there are in any given dataset, the more data is needed to see what is really happening. Therefore, it is essential that monitoring studies be done on a continual, long-term basis to help reduce the variability of the concentration data and systematically assess contaminate concentrations. For these reasons, continuing long-term monitoring programs are critical to protecting our environment and human health.

Given the complexity of the environment with the natural variability and uncertainty of the various environmental factors, the continuance of monitoring programs such as the SMWP and the NS&T are effective, essential tools for the stewardship of our environment. These monitoring programs allow researchers and decision makers to keep

abreast of changes in the concentration of chemicals in the environment and base their decisions on sound scientific data.

This thesis indicates, as expected, that many of the levels of the substances studied are decreasing. However, there are a few instances where the data points towards increasing levels of contaminants. For example, in the case of total DDT and total chlordane, both banned substances, increases are puzzling. A review of the recent literature suggests that decreasing trends in chemical concentrations are expected for chemicals that have been phased-out and ultimately banned. There could be numerous reasons for these unexpected increases. Most likely, both natural causes (i.e. flooding and sporadic heavy precipitation) and activities associated with human development (i.e. large scale land excavation) are stirring up old soil contaminants, such as lead, total DDT, and total chlordane; thus, increasing contaminated sediment runoff.

The most unexpected finding of this study was the indication that copper levels are on the rise. Probable rationales for the increase have been discussed previously. Increases across such a broad region bears further study.

RECOMMENDATIONS

The following are recommended changes to improve the monitoring process. First, transplanted mussels should be used at all locations to serve as control groups. Thus, differences in contaminate uptake between resident and transplant mussels at each site could be easily compared. Newer pesticides such as organophosphates and carbamates should be included in the monitoring programs (Stephenson, Martin, and Tjeerdema, 1995). Such expanded monitoring would provide a more complete picture of the potential risks of new chemicals. It is also recommended that sediment sampling be conducted at each mussel collection site to provide a broader background on the contaminants in the local environment. Specifically, site 404.0 in Moss Landing that showed increasing trends in DDT, chlordane, and PCBs, and sites 10.0 (Trinidad Head), 202.0 (Bodega Head), 414.0 (Pacific Grove), 443.0 (San Diego Bay) that showed increases in PCBs should be closely examined.

Following are a few recommendations to help lower the level of environmental contaminants. Due to the unexplained increases in copper levels it is again recommended that the causes for such increases be investigated. Further, it is recommended that the usage of copper for urban (i.e. automobile brake pads) and rural (fungicide) purposes be reduced. Better control of urban and agricultural sediment runoff is also suggested. Copper must be investigated to determine what potential negative health effects increased levels may have on the environment and humans in particular.

In future studies, the following are suggested: First, land use data should be reclassified and simplified, which would allow for a consistent land-use scheme that is

statistically valid and reproducible. Second, further research to investigate the population densities around sample sites and contamination levels in the California mussel would be beneficial. Third, river load and contaminate transport near the mussel sites should be investigated. Finally, a better understanding of mussel metabolism may be very helpful.

APPENDIX A. POTENTIAL HEALTH HAZARDS OF SPECIFIC CHEMICAL EXPOSURE

A general background on each of the six chemicals included in this study, a description of each chemical's uses and the possible risks to human and environmental health when exposed to a particular chemical are provided below. Knowing the potential sources of chemical contamination is important in speculating where contaminants possibly originate and what damage they can potentially do to humans and marine life, such as mussels.

Silver (Ag)

Introduction. Silver is a white, metallic chemical element found naturally in the environment. It is often combined with other elements such as sulfide, chloride, and nitrate (Agency for Toxic Substances and Disease Registry (ATSDR, 1990). Silver is commonly found as a by-product during the recovery of gold, copper, lead and zinc ores. Silver is used in the manufacture of coins, jewelry, silverware, and alloys due to its extreme malleability. It is also used in electronic equipment and in dental fillings. The photography industry is a heavy user of silver compounds. Silver bromide, AgBr, is the main salt used in photographic emulsions. Silver chloride, AgCl, is also used in photography and infrared spectroscopy. Silver iodide, AgI, is used in photography, medicine, and in seedling clouds to make rain. Silver nitrate, $AgNO_3$, is used as an antiseptic and in silver-plating and photography.

There are several uses of silver that promote the health of humans, including its use as a disinfectant in drinking water and water in swimming pools, as an antibacterial

agent, and as an ingredient in throat lozenges and chewing gum to aid people in their desire to stop smoking (ATSDR, 1990).

Silver is naturally released into the air through the processing of ores, cement manufacturing, and the burning of fossil fuels. Rain washes silver out of rocks and soil into the groundwater (ATSDR, 1990). Only trace amounts of silver are found naturally in drinking water (http://www.ci.boulder.co.us/environmentalaffairs/PACE).

Silver contamination usually occurs by the discharge of silver-containing solutions into the sanitary sewers. Silver contamination of water can be caused by disposal of industrial waste, including waste from manual and automated photographic processing businesses, metal plating, dental offices from x-ray film and fixers, and various laboratory methods such as biological stains and gel developing processes that contain silver (ATSDR, 1990). Silver recovered during the photo-processor fixing process is listed as a "heavy metal" pollutant by the Environmental Protection Agency (EPA) and is restricted to less than 5.0 milligrams per liter (mg/l) of waste water. Local authorities sometimes have much stricter limits---as low as 0.05 mg/l (Cooley and Raysin, 2001). In order to reduce the amount of silver discharged into sanitary sewers, silver-containing solutions are often processed through silver-recovery equipment or collected by hazardous waste specialists (Cooley and Raysin, 2001).

Potential Health Hazards. Human exposure to silver occurs when humans breath in airborne silver, swallow food or water with silver in it, touch, or otherwise come into contact with silver in such a way as that it is absorbed into the body (ATSDR, 1990). There is no evidence that mild exposure to silver is a health risk to humans (ATSDR,

1990). Silver is not known to be a carcinogen or to be the cause of any major health risk. However, exposure to high levels of silver has been shown to cause arygria, a permanent blue-gray discoloration of eyes, skin and mucous membranes that is cosmetically harmful (ATSDR, 1990). The EPA recommends that the concentration of silver in drinking water not exceed 0.10 milligrams per liter of water in order to avoid arygria. Spills of silver over 1000 pounds are required to be reported to the EPA (ATSDR, 1990).

Copper (Cu)

Introduction. Copper is a reddish-brown, malleable metallic element naturally occurring in the environment, plants and animals. Copper is an element essential to good health in all living things including people (ATSDR, 1990). In fact, copper is the mechanism by which mussels take up oxygen (Squire, personal communication, 2002).

Copper is found worldwide, including the United States, where it is heavily mined. It is used to make many items, including, but not limited to: wire, sheet metal, pipes, pennies, brake pads of automobiles, and boat paints (ATSDR, 1990). Copper sulfate, $CuSO_4$, is used in making pigments, germicides, and batteries.

Copper most commonly enters our environment through natural emissions. It is emitted naturally into air through wind blown dust and volcanic eruptions. Copper is emitted naturally into water through the weathering of soil (ATSDR, 1990). Copper may also enter the air through the following human activities: copper smelting and ore processing, fungicide use on plants or in lakes and ponds to control algae, and application

to wood, fabric, and leather. Copper can enter into water through industry discharges and sewage treatment plants (ATSDR, 1990).

Potential Health Hazards. Human exposure to copper occurs when humans breath in airborne copper, swallow food or water with copper in it (i.e. drinking water that passes through copper pipes), touch, or otherwise come into contact with copper in such a way as that it is absorbed into the body (ATSDR, 1990). There is no evidence that mild exposure to copper is a health risk to humans (ATSDR, 1990). Like silver, copper is not known to be a carcinogenetic. The International Agency for Research on Cancer (IARC) has concluded that copper is not classifiable as to human carcinogenicity (ATSDR, 1990).

However, long-term exposure to copper has been shown to cause: irritation of the nose, mouth and eyes, dizziness, headaches, and diarrhea. Exposure to high amounts of copper can cause: effects on the blood, and liver and kidney damage. Higher than normal levels of copper in drinking water can cause vomiting, diarrhea, stomach cramps, and nausea (ATSDR, 1990).

The EPA has set a treatment technique for copper in drinking water that includes an action level of 1.3 milligrams of copper per liter of water (1.3 mg/L). The EPA has also set a secondary maximum contaminant level (SMCL) of 1 mg/L of copper in drinking water. An SMCL is a non-enforceable drinking water standard based on taste, odor, or other aesthetic considerations. The EPA requires that spills or accidental releases into the environment of 5,000 pounds or more of copper be reported to the EPA. The Occupational Safety and Health Administration (OSHA) has set occupational

exposure limits of 0.1 milligram of copper per cubic meter or air (0.1) mg/m^3) as fumes and 1 mg/ m^3 of copper as dust and mists for a 8-hour workday, 40-hour workweek (ATSDR, 1990).

Lead (Pb)

Introduction. Lead is a naturally occurring bluish-gray, malleable metallic chemical element found in small amounts in the earth's crust. Lead does not break down, but its compounds can be altered by sunlight, air, and water. Lead can travel long distances in the air before settling to the ground. Once lead falls onto soil, it usually adheres to soil particles (ATSDR, 1990).

Many human activities introduce lead into our environment. Such activities include, but are not limited to: burning fossil fuels, mining, manufacturing, and the production of batteries, ammunition, metal products (alloys, solder and pipes), and devices to shield X-rays (ATSDR, 1990).

Lead is also introduced into our environment through usage of household products with high lead concentrations such as: leaded gasoline, lead-based paints, ceramic products, and caulking. These high lead products usually enter the environment through emission into the air and water. For example, a large portion of the high lead in city soils originated in older buildings painted with lead-based paint and varnishes made with poisonous, colorless lead acetate, $Pb(C_2H_3O_2)_2(3H_2O)$. In recent years, lead from gasoline, paints and ceramic products, caulking, and pipe solder has been dramatically reduced due in part to public awareness of the health risks associated with it and public

safety regulations implemented to restrict usage of products with high lead concentrations (ATSDR, 1990). Lead arsenate, $Pb_3(AsO_4)_2$, a very poisonous, colorless, crystalline compound is used as an insecticide and herbicide.

Potential Health Hazards. Human exposure to lead can occur in numerous ways, including the following: breathing lead air or dust, eating contaminated foods, drinking contaminated water, spending a significant amount of time in buildings where lead-based paints have been used and are deteriorating, spending a significant amount of time in a location where lead is used, using health-care products that contain lead, and using lead to create products such as stained glass (ATSDR, 1990).

Regardless of how it is exposed, the human body is sensitive to lead exposure. Lead can affect almost every organ and system in the body, especially the nervous system, kidneys, and reproductive system. At high levels of exposure, lead may decrease reaction time, cause weakness in fingers, wrists, or ankles, affect memory, and may cause anemia, a disorder of the blood (ATSDR, 1990).

There is no information regarding the affects of low levels of lead with regard to the above (ATSDR, 1990).

Based on animal studies, the Department of Health and Human Services (DHHS) has stated that lead acetate and lead phosphate most probably are carcinogens. However, there is no concrete evidence to prove lead's carcinogenicity in people (ATSDR, 1990).

A public health concern that has gotten much coverage in the media is the possible lead exposure of children, who are more vulnerable to lead poisoning than adults (ATSDR, 1990). Small children can be exposed to lead by eating lead-based paint chips

or by otherwise swallowing items that contain lead or by playing in contaminated soil. A child who swallows large amounts of lead may develop the severe symptoms listed above, including permanent brain damage. Even a small amount of a high lead based paint chip can result in a large amount of lead entering a child's body. As with adults, the less lead that a child is exposed to the less severe the effects on the child's blood and brain function. However, unlike adults, there is evidence that even at much lower levels of lead exposure, a child's mental and physical growth can be affected.

Lead has been found in at least 1,026 of 1,467 National Priorities List sites identified by the EPA (ATSDR, 1990). The following are a few of the actions that have been taken to reduce lead exposure: (1) EPA requirement that lead in air not exceed 1.5 micrograms per cubic meter (1.5 $\mu g/m^3$) over a 3 month period, (2) EPA limit of lead in drinking water to 15 μg per liter, (3) OHSA regulations for workers exposed to lead, (4) the U.S. government 1978 ban of lead-based paint for housing, (5) the Clean Air Act Amendments of 1990 that ban the sale of leaded gasoline, and (6) the Federal Hazardous Substance Act that bans children's products that contain hazardous amounts of lead.

Total chlordane

Introduction. Total chlordane is a manufactured chemical comprising of a mixture of pure chlordane and many related chemicals. Chlordane is not naturally found in the environment. It is a chlorinated, highly poisonous, volatile oil, $C_{10}H_6Cl_8$, formerly used as an insecticide from 1948 until 1988. It was widely used for controlling termites in

residential homes and on agricultural crops such as corn and citrus fruits, as well as on residential lawns and gardens (ATSDR, 1990).

With mounting concern over the potential damage to the environment and threat to human health, the EPA banned all agricultural uses of chlordane in 1983 and its use as a termiticide in 1988 (Shigenaka, 1990; O'Connor, 1996).

Environmental Concerns. Like lead, chlordane strongly adheres to soil particles. Although most chlordane will leave the soil through evaporation, it breaks down slowly and has the ability to remain in the soil for over 20 years (ATSDR, 1990). Through various routes of exposure, such as: contacting contaminated soil, breathing contaminated air, and eating contaminated microorganisms, fish or shellfish, it has the potential to accumulate in the tissues of fish, birds, and mammals (ATSDR, 1990).

Potential Health Hazards. Chlordane affects the central nervous system, the digestive system, and the liver in humans and animals. Breathing air containing high concentrations of chlordane or accidentally ingesting small amounts of chlordane have caused individuals to suffer from stomach cramps, diarrhea, vomiting, vision problems, weakness, confusion, jaundice, headaches and irritability (ATSDR, 1990). Large oral doses have cause convulsions and death in humans. Although mice given low doses of chlordane in food developed liver cancer, the IARC has not been able to classify chlordane as a carcinogenic to humans (ATSDR, 1990).

Federal Regulations. The EPA requires spills or releases of chlordane into the environment of 1 pound or greater to be reported to the EPA. The EPA has set drinking water limit of 2 ppb while the Food and Drug Administration (FDA) has imposed a

maximum of 300 ppb in most fruits and vegetables and a maximum of 100 ppb in animal fat and fish (ATSDR, 1990). A maximum level of 0.5 milligrams of chlordane per cubic meter (mg/m^3) in the workplace for an 8-hours day, 40 hour workweek has been established by the OSHA, the American Conference of Governmental Industrial Hygienists (ACGIH) and the National Institute for Occupational Health and Safety (NIOSH) (ATSDR, 1990).

Total DDTs

Introduction. In 1939, Paul Muller, who worked for Geigy Pharmaceutical of Switzerland discovered that DDT (1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane) could be used effectively as an inexpensive chlorinated organic insecticide. DDT is a white, crystalline solid with no odor or taste. For three decades after World War II, the world extensively manufactured and used DDT to combat the spread of insect carried diseases. DDT proved effective against mosquitoes, which spread malaria and against lice that carry typhus. Muller was awarded the Nobel Prize in medicine and physiology in 1948 for his discovery of such a revolutionary insecticide (ATSDR, 1990).

The World Health Organization has estimated that approximately 25 million lives were saved due to a reduction of insect spread diseases during the period of DDT's use. Based on initial studies, DDT was assumed to be safe, due to its relatively low toxicity to mammals (oral LD50 is 300 to 500 mg/kg) (Harrison, 1997). In the U.S. alone, approximately 675,000 tons of DDT was applied during the thirty-year span of its intensive agricultural and commercial usage (Harrison, 1997). In 1959, domestic DDT usage peaked at almost 80 million pounds (Harrison, 1997).

Environmental Concerns. DDT enters the environment when used as an insecticide. Airborne DDT lasts for only a short time, with a half-life of two days. DDT does not dissolve easily in water. Therefore, it attaches to soil particles and does not move quickly to underground water. DDT's half-life in soil is approximately two to 15 years. When DDT breaks down, some evaporates into the air and some is broken down by sunlight and/or by microorganisms in soil or surface water. Total DDT is the sum of concentrations of *ortho* and *para* forms of parent and metabolites 2,4'DDE; 4,4'DDE; 2,4'DDD; 4,4'DDD; 2,4'DDT; and 4,4'DDT (O'Connor, 1996). DDT in soil usually breaks down to form two similar chemicals DDE or DDD (ATSDR, 1994).

DDE (1,1-dichloro-2,2-bis(chlorophenyl) ethylene) was also used to kill pests, but its use has also been banned because it was found to inhibit calcium production, causing many birds to produce extremely thin eggshells that would lead to increased mortality. DDD (1,1-dichloro-2,2-bis(p-chlorophenyl) ethane) one form of DDD has been used medically to treat cancer of the adrenal gland.

So why is the use of DDT now banned in the United States? Problems associated with the use of DDT began to emerge in the late 1940s, when many species of insects developed resistance to DDT and its high toxicity toward fish was discovered. Rachel Carson's book, Silent Spring, warned of the danger of using DDT.

Most alarming was the discoveries of DDT's chemical stability (biological half-life of up to twenty years) and its fat solubility. It was discovered that mammals do not metabolize DDT very rapidly; rather, our bodies store it in fatty tissues. Therefore, if

ingestion continues at a steady rate, DDT builds up within the mammal's body (Harrison, 1997).

In 1972, except for public health emergencies, the use of DDT in the agricultural business was banned in the United States due to the fact that it presented undesirable risks to the environment and potential damage to human health (O'Connor, 1996). It is worthy to note that regardless of obvious ethical issues, the United States still manufactures and exports DDT for use in other countries. Former Vice President Al Gore stated in the introduction of Silent Spring, that exporting DDT "not only involves a readiness to profit by selling others a hazard we will not accept for ourselves; it also reflects an elemental failure to comprehend that the laws of science do not observe the boundaries of politics. Poisoning the food chain anywhere ultimately poisons the food chain everywhere."

Humans are exposed to low levels of DDT by consuming domestic foods, such as root and leafy vegetables, fatty meat, fish, and poultry. Humans can be exposed to higher levels of DDT when consuming imported foods from countries that still allow the use of DDT. In the United States, it is unlikely that humans will be exposed to DDT through air or water; as levels of DDT are generally low and of little concern except near waste sites and landfills that may contain higher levels of DDT (ATSDR, 1994).

Potential Health Hazards. DDT affects the nervous system. People who have accidentally swallowed large amounts of DDT became excitable and had tremors and seizures. These effects went away after the exposure stopped. No effects were seen in people who took small daily doses of DDT by capsule for 18 months. People who

worked with DDT for a long time had some reversible changes in the levels of liver enzymes (ATSDR, 1994).

In animals, short-term exposure to large amounts of DDT in food affected the nervous system. In animals, long-term exposure to DDT affected the liver. Animal studies suggest that short-term exposure to DDT in food may have a harmful effect on reproduction (ATSDR, 1994).

The DHHS has determined that DDT may reasonably be anticipated to be a human carcinogen. DHHS has not classified DDE and DDD, but the EPA has determined that they are probable human carcinogens (ATSDR, 1994). Liver cancer has been seen in animals that were fed DDT. Studies in DDT-exposed workers did not show increases in cancer. Laboratory tests can detect DDT, DDE, and DDD in fat, blood, urine, semen, and breast milk. These tests may show low, moderate, or excessive exposure to these compounds. These tests cannot show the exact amount of DDT, DDE, or DDD to which a person was exposed or tell if harmful effects will occur.

The EPA requires spills or releases of DDT into the environment of 1 pound or more be reported to them immediately (ATSDR, 1994). The FDA has set limits on DDT levels in most foods. The OSHA set an exposure limit of 1 milligram of DDT per cubic meter (1 mg/m^3) in workplace air for an 8-hour workday, 40-hour workweek. The NIOSH recommends an exposure limit of 0.5 mg/m^3 in workplace air over a 10-hour workday, 40-hour workweek (ATSDR, 1994).

Total PCBs

Introduction. PCBs are combinations of up to 209 individual chlorinated compounds (known as congeners). In this study total PCB is twice the sum of the concentrations of 18 congeners (PCB8, PCB18, PCB28, PCB44, PCB52, PCB66, PCB101, PCB105, PCB118, PCB128, PCB138, PCB153, PCB170, PCB180, PCB187, PCB195, PCB206, and PCB209 (O'Connor, 1996). There are no natural sources of PCBs. PCBs are a toxic, odorless, colorless to light yellow oily liquid, solid or vapor. The U.S. trade name Aroclor represented many commercial PCB combinations (ATSDR, 2000).

Large electrical transformers and other electrical equipment such as capacitors used PCBs as a coolant and a lubricant. PCBs provided good insulation and did not readily burn. Old fluorescent lighting ballasts and hydraulic oils commonly contained PCBs (ATSDR, 2000).

Environmental Concerns. In 1971, PCBs began being phased-out in the U.S. Further uses were banned in 1976 (O'Connor, 1996) and the manufacture of PCBs was ultimately stopped the following year due to evidence that they accumulated in the environment and had the potential to cause harmful health effects (ATSDR, 2000).

Despite the fact that PCBs have been banned in the U.S., they are still present in the environment. PCBs are found in the air, water, and soil because of inadvertent leaks and spills during transportation; leaks from old electrical transformers containing PCBs; leaks from hazardous waste sites; against the law or improper disposal of industrial

wastes and consumer products; and burning of some wastes in incinerators (ATSDR, 2000).

Since PCBs do not easily break down in the environment, they can travel long distances in the air and be deposited in areas far away from their original source. A small amount of PCBs can be water soluble, but most bond to organic particles, bottom sediments and soil (ATSDR, 2000).

Small aquatic organisms and fish can ingest PCBs in the water column and absorb the compounds in their tissues. The bioaccumulation of PCBs in aquatic organisms and marine mammals can reach levels that may be many thousands of times higher than in water (ATSDR, 2000).

Potential Health Hazards. Human exposure to large amounts of PCBs can cause skin conditions such as acne and rashes and possible liver damage.

Animals that ingested food containing large quantities of PCBs for short periods of time had mild liver damage and with some resulting in death. Animals that ate smaller amounts of PCBs in food over several weeks or months developed various kinds of health effects, including: anemia; acne-like skin conditions; and liver, stomach, and thyroid gland injuries. Other harmful effects of PCBs in mammals include: alteration of the immune system, behavioral changes, and harm to the reproductive system. PCBs are not known to cause birth defects (ATSDR, 2000).

A small number of studies of workers that were exposed to PCBs indicated that PCBs may be associated with certain kinds of cancer in humans, such as cancer of the liver and biliary tract. Study of rats that ate food containing high levels of PCBs for two

years showed development of liver cancer. The DHHS has determined that PCBs may reasonably be probable to be carcinogens. The EPA and the IARC have concluded that PCBs are most likely carcinogenic to humans (ATSDR, 2000).

Studies have shown that women who were exposed to moderately high levels of PCBs in the workplace or consumed large quantities of fish contaminated with PCBs had babies that weighed slightly less than babies born to women who did not have these PCB exposures. Babies born to women who ate PCB-contaminated fish also displayed nonstandard responses in tests of infant behavior. Some of the noted behaviors (i.e. problems with motor skills and a decrease in short-term memory) lasted for several years. In addition some studies suggest that the immune system was affected in children born to and nursed by mothers exposed to increased levels of PCBs. There is no indication of structural birth defects caused by exposure to PCBs or of health effects of PCBs in older children. Infants are exposed to PCBs in the highest quantities from breast milk. Transplacental transfers of PCBs have also been reported. However, in most cases, the benefits of breast-feeding offset any danger from exposure to PCBs in mother's milk (ATSDR, 2000).

There are tests available to measure levels of PCBs in blood, body fat, and breast milk; however, these are not routinely conducted. Most people generally have low levels of PCBs in their body because nearly everyone has been exposed to PCBs in some way. The aforementioned tests indicate if your PCB levels are high. Elevated PCB levels would indicate past exposure to above-normal levels of PCBs, but the tests cannot

determine when or how long you were exposed to PCBs or whether you will develop health effects from the exposure (ATSDR, 2000).

The EPA has set a limit of 0.0005 milligrams of PCBs per liter of drinking water. Any release of 1 pound or more of PCBs into the environment must be reported to the EPA. The FDA mandates that infant foods, eggs, milk and other dairy products, fish and shellfish (including mussels), poultry and red meat contain no more than 0.2-3 parts of PCBs per million parts of food. Numerous states have established fish and wildlife consumption notices for PCBs.

Table 6. List of the 24 Mussel Watch Sites Studied for Objective One. "R" represents Resident California mussels and "T" represents Transplanted California mussels.

Station Number	Station Name	Species collected	Name of Watershed(s)
2.0	Crescent City/STP Outfall	R	Smith River Plain
3.0	Crescent City/Control	R	Smith River Plain
10.0	Trinidad Head	R	Big Lagoon
100.0	Mad River Slough	T	Eureka Plain
103.0	Eureka Channel	T	Eureka Plain
104.5	Eureka STP/Control	T	Eureka Plain
202.0	Bodega Head	R	Bodega Head and Bodega Bay
336.0	J. Fitzgerald	R	Pacifica
404.0	Sandholdt Bridge	T	Neponset, Bolsa Neuva and Moro Cojo
414.0	Pacific Grove	R	Monterey Peninsula
430.0	Montana De Oro	R	Los Osos, Morro Bay and Point San Luis
434.0	Diablo Cove/South	R	Point San Luis
435.0	Intake Cove	R	Point San Luis
437.0	Point San Luis	R	Point San Luis and San Luis Obispo Ck.
443.0	Diablo Cove/South	T	Point San Luis
444.0	Intake Cove/Trans.	T	Point San Luis
445.0	San Luis Harbor	T	Point San Luis and San Luis Obispo Ck.
601.0	LA Harbor/National Steel	T	Lower Santa Monica Bay, Los Angeles, Carson, San Pedro, Los Angeles Harbor and Terminal Island
616.0	LA Harbor/ Consolidated Slip	T	Lower Santa Monica Bay, Los Angeles, Carson, San Pedro, Los Angeles Harbor and Terminal Island
662.0	Royal Palms	R	Lower Santa Monica Bay, Los Angeles, Carson, San Pedro, Los Angeles Harbor and Terminal Island
713.0	Huntington Harbour/ Edinger St.	T	Central (Split), Anaheim and East Coastal Plain
715.0	Huntington Harbour/ Warner Ave Bridge	T	Central (Split), Anaheim and East Coastal Plain
750.0	Oceanside	R	Lower Ysidora, Mission, Loma Alta and El Salto
894.0	SD Bay/Harbor Is/E Basin/Storm Dr	T	La Nacion, Chollas, Lindbergh, Point Loma, Paradise, El Toyan, Otay Valley, Telegraph, Coronado and San Diego Bay

Table 7. List of the 42 Mussel Watch Sites Studied for Objective Two. "R" represents Resident California mussels and "T" represents Transplanted California mussels.

Station Number	Station Name	Species collected	Name of Watershed(s)
2.0	Crescent City/STP Outfall	R	Smith River Plain
3.0	Crescent City/Control	R	Smith River Plain
10.0	Trinidad Head	R	Big Lagoon
100.0	Mad River Slough	T	Eureka Plain
103.0	Eureka Channel	T	Eureka Plain
104.5	Eureka STP/Control	T	Eureka Plain
202.0	Bodega Head	R	Bodega Head and Bodega Bay
205.0	Bodega Harbor/Spud Pt. Marina	T	Bodega Head and Bodega Bay
336.0	J. Fitzgerald	R	Pacifica
404.0	Sandholdt Bridge	T	Neponset, Bolsa Neuva and Moro Cojo
414.0	Pacific Grove	R	Monterey Peninsula
430.0	Montana De Oro	R	Los Osos, Morro Bay and Point San Luis
434.0	Diablo Cove/South	R	Point San Luis
435.0	Intake Cove	R	Point San Luis
437.0	Point San Luis	R	Point San Luis and San Luis Obispo Creek
443.0	Diablo Cove/South	T	Point San Luis
444.0	Intake Cove/Transplant	T	Point San Luis
445.0	San Luis Harbor/Transplant	T	Point San Luis and San Luis Obispo Ck.
601.0	LA Harbor/National Steel	T	Lower Santa Monica Bay, Los Angeles, Carson, San Pedro, Los Angeles Harbor and Terminal Island
616.0	LA Harbor/Consolidated Slip	T	Lower Santa Monica Bay, Los Angeles, Carson, San Pedro, Los Angeles Harbor and Terminal Island
618.0	LA Harbor/Angels Gate	R	Lower Santa Monica Bay, Los Angeles, Carson, San Pedro, Los Angeles Harbor and Terminal Island
648.0	Malibu	R	Corral Canyon, Monte Nido, Carbon Canyon and Las Flores Canyon

Station Number	Station Name	Species collected	Name of Watershed(s)
650.0	Santa Monica	R	Upper Santa Monica Bay and Culver City
662.0	Royal Palms	R	Lower Santa Monica Bay, Los Angeles, Carson, San Pedro, Los Angeles Harbor and Terminal Island
708.0	Anaheim Bay/Navy Marsh	T	Central (Split), Anaheim, and East Coastal Plain
713.0	Huntington Harbour/Edinger Street	T	Central (Split), Anaheim and East Coastal Plain
715.0	Huntington Harbour/Warner Ave Bridge	T	Central (Split), Anaheim and East Coastal Plain
723.4	Newport Bay/Turning Basin	T	East Coastal Plain and Newport Bay
724.0	Newport Bay/Highway 1 Bridge	T	East Coastal Plain and Newport Bay
725.0	Newport Bay/Crows Nest	T	East Coastal Plain and Newport Bay
726.4	Newport Bay/Rhine Channel/End	T	East Coastal Plain and Newport Bay
744.1	San Onofre 1	T	San Mateo Canyon and San Onofre Valley
744.3	San Onofre 3	T	San Mateo Canyon and San Onofre Valley
744.4	San Onofre 4	T	San Mateo Canyon and San Onofre Valley
744.5	San Onofre 5	T	San Mateo Canyon and San Onofre Valley
744.6	San Onofre 6	T	San Onofre Valley and Las Pulgas
750.0	Oceanside	R	Lower Ysidora, Mission, Loma Alta and El Salto
883.6	San Diego Bay/7th Street Channel	T	La Nacion, Chollas, Lindbergh, Point Loma, Paradise, El Toyan, Otay Valley, Telegraph, Coronado and San Diego Bay
883.8	San Diego Bay/Switzer Creek	T	La Nacion, Chollas, Lindbergh, Point Loma, Paradise, El Toyan, Otay Valley, Telegraph, Coronado and San Diego Bay

Station Number	Station Name	Species collected	Name of Watershed(s)
887.0	San Diego Bay/Evans Street	T	La Nacion, Chollas, Lindbergh, Point Loma, Paradise, El Toyan, Otay Valley, Telegraph, Coronado and San Diego Bay
894.0	SD Bay/Harbor Is/E Basin/Storm Dr	T	La Nacion, Chollas, Lindbergh, Point Loma, Paradise, El Toyan, Otay Valley, Telegraph, Coronado and San Diego Bay Telegraph Coronado and San Diego Bay
899.0	San Diego Bay/Shelter Is/Fshg Pier	T	La Nacion, Chollas, Lindbergh, Point Loma, Paradise, El Toyan, Otay Valley, Telegraph, Coronado and San Diego Bay

Table 8. Regression Results of long-term trends in silver (Ag) at selected Mussel Watch sites in California. Results from this study are indicated with a (Larson) after the station name and those from Stephenson et al. are on the second line for each station. Relationships that had higher R2 values when regressed as higher order polynomial or exponential equations than as linear relationships are shown as 'Poly' and 'Exp', respectively. 'NA' indicates a p value was not applicable in these sites. 'NR' means no relationship over time was statistically detected. R=resident California mussel. T=transplant California mussel. 'DL' indicates more than 80% of the values at a site were below the detection limit for the element. Detection limits for silver was 0.002 ppm dry weight. Not significant, 'NS', increasing, 'I', and decreasing, 'D', trends were identified.

Mussel Type	Site No.	Site Location	n	Slope ppb/yr	Ag R2	Ag p-value	Not significant, Increasing (I) or Decreasing (D) Trend
R	2	Crescent City/STP Outfall (Larson)	11		NR		NS
R	2	(Stephenson)	8	Exp	0.372	NA	D
R	3	Crescent City/Control (Larson)	13		NR		NS
R	3	(Stephenson)	8		NR		NS
R	10	Trinidad Head (Larson)	21	-0.002	0.293	0.0093	D
R	10	(Stephenson)	14		NR		NS
R	202	Bodega Head (Larson)	33	-0.004	0.383	0.0001	D
R	202	(Stephenson)	23	-0.005	0.179	<.05	D
R	414	Pacific Grove (Larson)	25	-0.074	0.529	0.0000	D
R	414	(Stephenson)	15	-0.147	0.564	<.001	D
R	430	Montana De Oro (Larson)	28	-0.016	0.323	0.0005	D
R	430	(Stephenson)	23	-0.020	0.337	<.01	D
T	616	LA Harbor/Consolidated Slip (Larson)	17		NR		NS
T	616	(Stephenson)	6		NR		NS
R	662	Royal Palms (Larson)	24	-0.201	0.333	0.0032	D
R	662	(Stephenson)	14		NR		NS
T	715	Huntington Harbour/Warner Ave Brdg	17		NR		NS
T	715	(Stephenson)	7	-0.008	0.453	<.10	D
R	750	Oceanside (Larson)	23	-0.040	0.469	0.0003	D
R	750	(Stephenson)	15	Poly	0.885	NA	D
T	894	SD Bay/Harbor Is/E Basin/Storm Dr	10	Poly	0.744	0.0329	D
T	894	(Stephenson)	6	0.138	0.745	<.05	I

Table 9. Regression Results of long-term trends in lead (Pb) at selected Mussel Watch sites in California. Results from this study are indicated with a (Larson) after the station name and those from Stephenson et al. are on the second line for each station. Relationships that had higher R2 values when regressed as higher order polynomial or exponential equations than as linear relationships are shown as 'Poly' and 'Exp', respectively. 'NA' indicates a p value was not applicable in these sites. 'NR' means no relationship over time was statistically detected. R=resident California mussel. T=transplant California mussel. 'DL' indicates more than 80% of the values at a site were below the detection limit for the element. Detection limits for lead was 0.03 ppm dry weight. Not significant, 'NS', increasing, 'I', and decreasing, 'D', trends were identified.

Mussel Type	Site No.	Site Location	n	Slope ppb/yr	Pb R2	Pb p-value	Not significant, Increasing (I) or Decreasing (D) Trend
R	2	Crescent City/STP Outfall (Larson)	11		NR		NS
R	2	(Stephenson)	8		NR		NS
R	3	Crescent City/Control (Larson)	13		NR		NS
R	3	(Stephenson)	8		NR		NS
R	10	Trinidad Head (Larson)	21		NR		NS
R	10	(Stephenson)	14		NR		NS
R	202	Bodega Head (Larson)	33	-0.022	0.147	0.0278	D
R	202	(Stephenson)	23	-0.048	0.172	<.05	D
R	414	Pacific Grove (Larson)	25	-0.455	0.216	0.0194	D
R	414	(Stephenson)	15		NR		NS
R	430	Montana De Oro (Larson)	28		NR		NS
R	430	(Stephenson)	23	-0.076	0.169	<.10	D
T	616	LA Harbor/Consolidated Slip (Larson)	17	-1.35	0.835	0.0000	D
T	616	(Stephenson)	6	-2.17	0.625	<.10	D
R	662	Royal Palms (Larson)	24	-0.612	0.776	0.0000	D
R	662	(Stephenson)	14	-0.894	0.693	<.001	D
T	715	Huntington Harbour/Warner Ave Brdg	17	-1.39	0.727	0.0000	D
T	715	(Stephenson)	7		NR		NS
R	750	Oceanside (Larson)	23	-0.091	0.627	0.0000	D
R	750	(Stephenson)	15	Poly	0.703	NA	D
T	894	SD Bay/Harbor Is/E Basin/Storm Dr	10		NR		NS
T	894	(Stephenson)	6		NR		NS

Table 10. Regression Results of long-term trends in copper (Cu) at selected Mussel Watch sites in California. Results from this study are indicated with a (Larson) after the station name and those from Stephenson et al. are on the second line for each station. Relationships that had higher R2 values when regressed as higher order polynomial or exponential equations than as linear relationships are shown as 'Poly' and 'Exp', respectively. 'NA' indicates a p value was not applicable in these sites. 'NR' means no relationship over time was statistically detected. R=resident California mussel. T=transplant California mussel. 'DL' indicates more than 80% of the values at a site were below the detection limit for the element. Detection limits for copper was 0.003 ppm dry weight. Not significant, 'NS', increasing, 'I', and decreasing, 'D', trends were identified.

Mussel Type	Site No.	Site Location	n	Slope ppb/yr	Cu R2	Cu p-value	Not significant, Increasing (I) or Decreasing (D) Trend
R	2	Crescent City/STP Outfall (Larson)	11	0.413	0.418	0.0315	I
R	2	(Stephenson)	8		NR		NS
R	3	Crescent City/Control (Larson)	13	Poly	0.601	0.0222	D
R	3	(Stephenson)	8		NR		NS
R	10	Trinidad Head (Larson)	21		NR		NS
R	10	(Stephenson)	14		NR		NS
R	202	Bodega Head (Larson)	33	Exp	0.143	0.0301	I
R	202	(Stephenson)	23	0.116	0.149	<.10	I
R	414	Pacific Grove (Larson)	25	Exp	0.159	0.0483	I
R	414	(Stephenson)	15	0.228	0.314	<.05	I
R	430	Montana De Oro (Larson)	28	0.194	0.171	0.0151	I
R	430	(Stephenson)	23	0.251	0.196	<.05	I
T	616	LA Harbor/Consolidated Slip (Larson)	17		NR		NS
T	616	(Stephenson)	6		NR		NS
R	662	Royal Palms (Larson)	24		NR		NS
R	662	(Stephenson)	14		NR		NS
T	715	Huntington Harbour/Warner Ave Brdg	17	0.434	0.226	0.0537	I
T	715	(Stephenson)	7	1.58	0.466	<.10	I
R	750	Oceanside (Larson)	23		NR		NS
R	750	(Stephenson)	15		NR		NS
T	894	SD Bay/Harbor Is/E Basin/Storm Dr	10	Exp	0.458	0.0316	I
T	894	(Stephenson)	6	3.54	0.917	<.01	I

Table 11. Regression Results of long-term trends in total chlordane at selected Mussel Watch sites in California. Results from this study are indicated with a (Larson) after the station name and those from Stephenson et al. are on the second line for each station. Relationships that had higher R2 values when regressed as higher order polynomial or exponential equations than as linear relationships are shown as 'Poly' and 'Exp', respectively. 'NA' indicates a p value was not applicable in these sites. 'NR' means no relationship over time was statistically detected. 'NTANAL' indicated the compound was not analyzed at that station. R=resident California mussel. T=transplant California mussel. 'DL' indicates more than 80% of the values at a site were below the detection limit for the element. Detection limits for total chlordane was 1.0 ppb dry weight. Not significant, 'NS', increasing, 'I', and decreasing, 'D', trends were identified.

Mussel Type	Site No.	Site Location	n	Slope ppb/yr	Total Chlordane R2	Total Chlordane p-value	Not significant, Increasing (I) or Decreasing (D) Trend
R	2	Crescent City/STP Outfall (Larson)	11		NR		NS
R	2	(Stephenson)	10		NR		NS
R	3	Crescent City/Control (Larson)	14		NR		NS
R	3	(Stephenson)	10		NR		NS
R	10	Trinidad Head (Larson)	23	Poly	0.470	0.0202	D
R	10	(Stephenson)	17		NR		NS
T	100	Mad River Slough (Larson)	13		NR		NS
T	100	(Stephenson)	7	Poly	0.973	NA	D
T	103	Eureka Channel (Larson)	10	Poly	0.919	0.012	I
T	103	(Stephenson)	7		NTANAL		
T	104.5	Eureka STP/Control (Larson)	10		NR		NS
T	104.5	(Stephenson)	9		NR		NS
R	202	Bodega Head (Larson)	28	-0.600	0.411	0.0007	D
R	202	(Stephenson)	20	Poly	0.679	NA	D
R	336	J. Fitzgerald (Larson)	10	-0.002	0.51	0.111	NS
R	336	(Stephenson)	7		NTANAL		
T	404	Sandholdt Bridge (Larson)	14	Poly	0.913	0.0000	D
T	404	(Stephenson)	23	Exp	0.722	NA	D
R	414	Pacific Grove (Larson)	24	-0.984	0.490	0.0009	D
R	414	(Stephenson)	15	Poly	0.922	NA	D
T	601	LA Harbor/National Steel (Larson)	16	-4.83	0.389	0.0227	D
T	601	(Stephenson)	11		NR		NS
T	616	LA Harbor/Consolidated Slip (Larson)	18	-6.91	0.540	0.0012	D
T	616	(Stephenson)	12		NR		NS
R	662	Royal Palms (Larson)	25	-1.99	0.453	0.0011	D
R	662	(Stephenson)	17	-3.9	0.528	0.007	D
T	713	Huntington Harbour/Edinger Street (Larson)	14	-7.46	0.621	0.0008	D
T	713	(Stephenson)	7		NR		NS
T	715	Huntington Harbour/Warner Ave Brdg (Larson)	17	-15.0	0.701	0.0000	D
T	715	(Stephenson)	9		NR		NS
T	724	Newport Bay/Highway 1 Bridge (Larson)	16	-17.3	0.688	0.0001	D
T	724	(Stephenson)	10	-26.6	0.563	0.012	D
T	725	Newport Bay/Crows Nest (Larson)	19	-19.1	0.726	0.0000	D
T	725	(Stephenson)	11	-30.5	0.621	0.012	D
T	726.4	Newport Bay/Rhine Channel/End (Larson)	14	-11.3	0.657	0.0008	D
T	726.4	(Stephenson)	8		NR		NS
T	744.6	San Onofre 6 (Larson)	7		NR		NS
T	744.6	(Stephenson)	7		NR		NS
R	750	Oceanside (Larson)	24	-2.53	0.493	0.0008	D
R	750	(Stephenson)	17	Exp	0.715	NA	D
T	894	SD Bay/Harbor Is/E Basin/Storm Dr (Larson)	12		NR		NS
T	894	(Stephenson)	12		NR		NS

Table 12. Regression Results of long-term trends in total DDT at selected Mussel Watch sites in California. Results from this study are indicated with a (Larson) after the station name and those from Stephenson et al. are on the second line for each station. Relationships that had higher R2 values when regressed as higher order polynomial or exponential equations than as linear relationships are shown as 'Poly' and 'Exp', respectively. 'NA' indicates a p value was not applicable in these sites. 'NR' means no relationship over time was statistically detected. 'NTANAL' indicated the compound was not analyzed at that station. R=resident California mussel. T=transplant California mussel. 'DL' indicates more than 80% of the values at a site were below the detection limit for the element. Detection limits for total DDT was 5.0 ppb dry weight. Not significant, 'NS', increasing, 'I', and decreasing, 'D', trends were identified.

Mussel Type	Site No.	Site Location	n	Slope ppb/yr	Total DDT R2	Total DDT p-value	Not significant, Increasing (I) or Decreasing (D) Trend
R	2	Crescent City/STP Outfall (Larson)	11		NR		NS
R	2	(Stephenson)	10		NR		NS
R	3	Crescent City/Control (Larson)	14		NR		NS
R	3	(Stephenson)	10		NR		NS
R	10	Trinidad Head (Larson)	23	-0.407	0.437	0.0006	D
R	10	(Stephenson)	17	Poly	0.545	NA	D
T	100	Mad River Slough (Larson)	13	Poly	0.821	0.0258	D
T	100	(Stephenson)	7		DL		
T	103	Eureka Channel (Larson)	10	Poly	0.825	0.0542	D
T	103	(Stephenson)	7		NTANAL		
T	104.5	Eureka STP/Control (Larson)	10		NR		NS
T	104.5	(Stephenson)	9		DL		
R	202	Bodega Head (Larson)	28		NR		NS
R	202	(Stephenson)	20		NR		NS
R	336	J. Fitzgerald (Larson)	10		NR		NS
R	336	(Stephenson)	7		NR		NS
T	404	Sandholdt Bridge (Larson)	14	Poly	0.553	0.0383	I
T	404	(Stephenson)	23	Poly	0.603	NA	D
R	414	Pacific Grove (Larson)	24		NR		NS
R	414	(Stephenson)	15		NR		NS
T	601	LA Harbor/National Steel (Larson)	16	-55.0	0.559	0.0014	D
T	601	(Stephenson)	11	-99.2	0.449	0.048	D
T	616	LA Harbor/Consolidated Slip (Larson)	18	-76.7	0.507	0.0009	D
T	616	(Stephenson)	12	Poly	0.924	NA	D
R	662	Royal Palms (Larson)	25	-31.4	0.209	0.0216	D
R	662	(Stephenson)	17		NR		NS
T	713	Huntington Harbour/Edinger Street (Larson)	14	-31.8	0.435	0.0102	D
T	713	(Stephenson)	7	Poly	0.735	NA	D
T	715	Huntington Harbour/Warner Ave Brdg (Larson)	17	-46.3	0.411	0.0056	D
T	715	(Stephenson)	9	-97.8	0.345	0.096	D
T	724	Newport Bay/Highway 1 Bridge (Larson)	16	-143	0.592	0.0005	D
T	724	(Stephenson)	10	Poly	0.733	NA	D
T	725	Newport Bay/Crows Nest (Larson)	19	-88.2	0.662	0.0001	D
T	725	(Stephenson)	11	Poly	0.811	NA	D
T	726.4	Newport Bay/Rhine Channel/End (Larson)	14	-59.9	0.402	0.02	D
T	726.4	(Stephenson)	8		NR		NS
T	744.6	San Onofre 6 (Larson)	7		NR		NS
T	744.6	(Stephenson)	7		NR		NS
R	750	Oceanside (Larson)	24	-8.55	0.161	0.0575	D
R	750	(Stephenson)	17		NR		NS
T	894	SD Bay/Harbor Is/E Basin/Storm Dr (Larson)	12		NR		NS
T	894	(Stephenson)	12		NR		NS

Table 13. Regression Results of long-term trends in total PCBs at selected Mussel Watch sites in California. Results from this study are indicated with a (Larson) after the station name and those from Stephenson et al. are on the second line for each station. Relationships that had higher R2 values when regressed as higher order polynomial or exponential equations than as linear relationships are shown as 'Poly' and 'Exp', respectively. 'NA' indicates a p value was not applicable in these sites. 'NR' means no relationship over time was statistically detected. 'NTANAL' indicated the compound was not analyzed at that station. R=resident California mussel. T=transplant California mussel. 'DL' indicates more than 80% of the values at a site were below the detection limit for the element. Detection limits for total PCBs was 1.0 ppb dry weight. Not significant, 'NS', increasing, 'I', and decreasing, 'D', trends were identified.

Mussel Type	Site No.	Site Location	n	Slope ppb/yr	Total PCBs R2	Total PCBs p-value	Not significant, Increasing (I) or Decreasing (D) Trend
R	2	Crescent City/STP Outfall (Larson)	11		NR		NS
R	2	(Stephenson)	10		DL		
R	3	Crescent City/Control (Larson)	14		NR		NS
R	3	(Stephenson)	10		DL		
R	10	Trinidad Head (Larson)	23	Poly	0.482	0.0014	I
R	10	(Stephenson)	17		DL		
T	100	Mad River Slough (Larson)	13		NR		NS
T	100	(Stephenson)	7		DL		
T	103	Eureka Channel (Larson)	10	Poly	0.855	0.0153	D
T	103	(Stephenson)	7		DL		
T	104.5	Eureka STP/Control (Larson)	10		NR		NS
T	104.5	(Stephenson)	9		DL		
R	202	Bodega Head (Larson)	28	Poly	0.362	0.0118	I
R	202	(Stephenson)	20		DL		
R	336	J. Fitzgerald (Larson)	10		NR		NS
R	336	(Stephenson)	7		DL		
T	404	Sandholdt Bridge (Larson)	14	Poly	0.523	0.0247	I
T	404	(Stephenson)	23		NR		NS
R	414	Pacific Grove (Larson)	24	Poly	0.305	0.0264	I
R	414	(Stephenson)	15		DL		
R	430	Montana De Oro (Larson)	28		DL		
R	430	(Stephenson)	28		DL		
R	434	Diablo Cove/South (Larson)	19		DL		
R	434	(Stephenson)	19		DL		
R	435	Intake Cove (Larson)	19		NR		NS
R	435	(Stephenson)	20		DL		
R	437	Point San Luis (Larson)	18		NR		NS
R	437	(Stephenson)	18		NR		NS
T	443	Diablo Cove/South/Transplant (Larson)	16	Poly	0.542	0.0062	I
T	443	(Stephenson)	15		DL		
T	444	Intake Cove/Transplant (Larson)	17	-3.30	0.252	0.04	D
T	444	(Stephenson)	17		DL		
T	445	San Luis Harbor/Transplant (Larson)	18		NR		NS
T	445	(Stephenson)	18		NR		NS
T	601	LA Harbor/National Steel (Larson)	16	-108	0.314	0.024	I
T	601	(Stephenson)	11		NR		NS
T	616	LA Harbor/Consolidated Slip (Larson)	18	-39.2	0.608	0.0001	D
T	616	(Stephenson)	12		NR		NS
R	662	Royal Palms (Larson)	25	-10.9	0.423	0.0004	D
R	662	(Stephenson)	17	Poly	0.618	NA	D
T	713	Huntington Harbour/Edinger Street (Larson)	14	-21.8	0.490	0.0053	D
T	713	(Stephenson)	7		NR		NS
T	715	Huntington Harbour/Warner Ave Brdg (Larson)	17	-28.8	0.543	0.0007	D
T	715	(Stephenson)	9		NR		NS
T	724	Newport Bay/Highway 1 Bridge (Larson)	16	-24.2	0.452	0.0043	D
T	724	(Stephenson)	10		NR		NS
T	725	Newport Bay/Crows Nest (Larson)	19	-227	0.782	0.0000	D
T	725	(Stephenson)	11	-183	0.365	0.049	D
T	726.4	Newport Bay/Rhine Channel/End (Larson)	14		NR		NS
T	726.4	(Stephenson)	8		NR		NS
T	744.6	San Onofre 6 (Larson)	7	9.19	0.668	0.0247	I
T	744.6	(Stephenson)	7		DL		
R	750	Oceanside (Larson)	24	-4.09	0.327	0.0105	D
R	750	(Stephenson)	17	-4.7	0.252	0.04	D
T	894	SD Bay/Harbor Is/E Basin/Storm Dr (Larson)	12	Poly	0.837	0.0016	I
T	894	(Stephenson)	12		NR		NS

Table 14. Chemical Contaminant Data for Each of the 42 Mussel Watch Sites. Trace metal and organic data are in ppm and ppb respectively.

SN	Site Name	Ag	Cu	Pb	T. Chlordane	T. DDT	T. PCB
2.0	Crescent City	3.82	12.63	2.47	18.08	33.25	21.00
3.0	Crescent City	.03	9.32	.93	1.26	2.06	3.71
10.0	Trinidad Head	.02	7.88	1.09	2.30	2.28	5.00
100.0	Mad River Slough	.05	10.35	1.63	3.54	1.40	23.40
103.0	Eureka Channel	.03	10.67	1.87	2.75	3.68	6.75
104.5	Eureka STP	.02	10.57	1.59	.70	3.25	.00
202.0	Bodega Head	.06	7.94	.96	2.03	6.88	12.29
205.0	Bodega Harbor	.02	16.05	2.26	4.31	13.33	87.66
336.0	J. Fitzgerald	.42	6.40	1.14	1.98	6.98	66.75
404.0	Sandholdt Bridge	.03	11.39	1.84	84.50	2456.88	231.69
414.0	Pacific Grove	.19	7.75	3.14	5.85	33.70	14.27
430.0	Montana De Oro	.07	6.52	.69	2.65	12.45	9.25
434.0	Diablo Cove	.20	6.46	1.20	.00	.00	12.40
435.0	Intake Cove	.17	6.47	.98	.00	.00	7.80
437.0	Point San Luis	.18	6.89	1.47	.00	.00	44.76
443.0	Diablo Cove	.11	6.56	.98	.00	.00	18.18
444.0	Intake Cove	.14	7.77	1.05	.00	.00	11.06
445.0	San Luis Harbor	.07	8.20	1.06	.00	.00	184.66
601.0	LA Harbor	.09	26.28	8.32	60.27	526.58	833.24
616.0	LA Harbor	.10	20.19	12.10	89.61	509.39	644.48
618.0	LA Harbor	.06	11.10	1.53	39.93	1162.27	519.70
648.0	Malibu	.85	7.95	1.02	18.03	95.00	52.15
650.0	Santa Monica	2.65	12.33	3.47	54.97	244.84	288.61
662.0	Royal Palms	3.35	9.08	2.30	13.65	722.39	120.15
708.0	Anaheim Bay	.66	11.33	4.03	48.32	394.04	192.22
713.0	Huntington Harbour	.06	15.67	6.07	81.93	357.02	184.89
715.0	Huntington Harbour	.10	16.50	11.33	114.91	385.98	256.03
723.4	Newport Bay	.10	38.31	6.97	72.25	313.81	248.43
724.0	Newport Bay	.06	25.82	4.32	106.53	765.79	226.00
725.0	Newport Bay	.05	73.95	6.98	80.15	609.57	1259.37
726.4	Newport Bay	.07	119.92	7.18	82.16	458.82	1057.39
744.1	San Onofre 1	1.01	7.53	1.19	8.18	105.80	29.00
744.3	San Onofre 3	.60	8.59	1.41	8.45	114.63	23.98
744.4	San Onofre 4	.84	7.54	1.46	9.30	101.75	21.30
744.5	San Onofre 5	.73	7.22	1.34	7.58	96.03	23.80
744.6	San Onofre 6	.46	7.39	1.24	7.40	91.86	21.70
750.0	Oceanside	.25	8.50	.76	9.28	71.46	32.75
883.6	San Diego Bay	.45	60.78	8.24	28.55	44.25	630.05
883.8	San Diego Bay	.21	34.50	8.22	34.58	31.23	509.83
887.0	San Diego Bay	.26	39.64	6.69	34.76	49.94	801.20
894.0	San Diego Bay	1.27	58.68	21.92	63.98	215.88	19093.60
899.0	San Diego Bay	.09	15.63	4.69	19.30	49.13	436.00

Table 15. Definitions and descriptions of independent variable sets used in the stepwise regressions for Objective 2.

Group Name	Variables in Group	Group Description
R1T2	1) Resident sites 2) Transplant sites	Each site had either resident or transplanted mussels
AVGRFX	1) Average rainfall	Average rainfall for five months at each site
Developed	1) "Commercial, Industrial, and Transportation" class 2) "Low Intensity Residential" class 3) "High Intensity Residential" class 4) "Urban/Recreational Grasses" class	Grouped four National Land Cover Designation (NLCD) variables that covaried together and are in the same urbanized land-use category. Summed total square meters of land cover for each designation per watershed.
Open Space	1) "Deciduous Forest" class 2) "Evergreen Forest" class 3) "Mixed Forest" class 4) "Woody Wetlands" class 5) "Transitional" class 6) "Pasture" class	Grouped six NLCD variables that covaried together and are in the same type of land-use category. Summed total square meters of land cover for each designation per watershed.
Agricultural -row crop*	1) "Row Crops" class	Summed total square meters of land cover for row crops designation per watershed.
Agricultural -no row crop*	1) "Fallow" class 2) "Orchards, Vineyards, and other non-natural woody" class	Grouped two variables that covaried together and are in the same type of land-use category. Summed total square meters of land cover for each designation per watershed.
Grasslands	1) "Grasslands/Herbaceous" class	Summed total square meters of land cover for grasslands designation per watershed.
Shrub	1) "Shrubland" class	Summed total square meters of land cover for shrubland designation per watershed.
Water	1) "Open Water" class	Summed total square meters of land cover for open water designation per watershed.

* Natural log transformed, ln(x+1), to improve normality for the analysis.

Table 16. Land-use Data in Square Meters for Each of the 42 Mussel Watch Sites. Please note site names have been abbreviated.

SN	Site Name	Developed	Open Space	LN Row Crops	LN Fallow /Orchard	Grass	Shrub	Water
2.0	Crescent City	7707600.0	192317400.0	14.6	.0	31083300.0	1238400.0	19082700.0
3.0	Crescent City	7707600.0	192317400.0	14.6	.0	31083300.0	1238400.0	19082700.0
10.0	Trinidad Head	2488500.0	192064500.0	8.4	.0	9063000.0	3611700.0	9943200.0
100.0	Mad River Slough	36340200.0	386797500.0	13.2	.0	50075100.0	17883900.0	69377400.0
103.0	Eureka Channel	36340200.0	386797500.0	13.2	.0	50075100.0	17883900.0	69377400.0
104.5	Eureka STP	36340200.0	386797500.0	13.2	.0	50075100.0	17883900.0	69377400.0
202.0	Bodega Head	1092600.0	2808900.0	7.5	7.5	16127100.0	600300.0	4350600.0
205.0	Bodega Harbor	1092600.0	2808900.0	7.5	7.5	16127100.0	600300.0	4350600.0
336.0	J. Fitzgerald	12354300.0	20139300.0	.0	14.2	20661300.0	20834100.0	837900.0
404.0	Sandholdt Bridge	55084500.0	92525400.0	19.1	13.6	76662900.0	34458300.0	3357000.0
414.0	Pacific Grove	62969400.0	68099400.0	13.4	20.6	98136900.0	73111500.0	633600.0
430.0	Montana De Oro	7861500.0	68311800.0	15.2	22.9	55904400.0	44819100.0	6763500.0
434.0	Diablo Cove	456300.0	48672900.0	7.5	9.3	26450100.0	34463700.0	877500.0
435.0	Intake Cove	456300.0	48672900.0	7.5	9.3	26450100.0	34463700.0	877500.0
437.0	Point San Luis	21845700.0	110353500.0	15.6	24.8	115494300.0	73501200.0	1548900.0
443.0	Diablo Cove	456300.0	48672900.0	7.5	9.3	26450100.0	34463700.0	877500.0
444.0	Intake Cove	456300.0	48672900.0	7.5	9.3	26450100.0	34463700.0	877500.0
445.0	San Luis Harbor	21845700.0	110353500.0	15.6	24.8	115494300.0	73501200.0	1548900.0
601.0	LA Harbor	676414800.0	24001200.0	12.8	18.9	25376400.0	82965600.0	23580900.0
616.0	LA Harbor	676414800.0	24001200.0	12.8	18.9	25376400.0	82965600.0	23580900.0
618.0	LA Harbor	676414800.0	24001200.0	12.8	18.9	25376400.0	82965600.0	23580900.0
648.0	Malibu	5045400.0	11703600.0	8.4	9.9	5265000.0	68486400.0	1565100.0
650.0	Santa Monica	106181100.0	17205300.0	9.5	9.2	7439400.0	77094900.0	1806300.0
662.0	Royal Palms	676414800.0	24001200.0	12.8	18.9	25376400.0	82965600.0	23580900.0
708.0	Anaheim Bay	875258100.0	79018200.0	16.0	23.3	95536800.0	180282600.0	7146900.0
713.0	Huntington Harbour	875258100.0	79018200.0	16.0	23.3	95536800.0	180282600.0	7146900.0
715.0	Huntington Harbour	875258100.0	79018200.0	16.0	23.3	95536800.0	180282600.0	7146900.0
723.4	Newport Bay	474282900.0	61881300.0	16.0	22.8	71112600.0	138673800.0	8853300.0
724.0	Newport Bay	474282900.0	61881300.0	16.0	22.8	71112600.0	138673800.0	8853300.0
725.0	Newport Bay	474282900.0	61881300.0	16.0	22.8	71112600.0	138673800.0	8853300.0
726.4	Newport Bay	474282900.0	61881300.0	16.0	22.8	71112600.0	138673800.0	8853300.0
744.1	San Onofre 1	7314300.0	89084700.0	13.8	17.2	38104200.0	353460600.0	704700.0
744.3	San Onofre 3	7314300.0	89084700.0	13.8	17.2	38104200.0	353460600.0	704700.0
744.4	San Onofre 4	7314300.0	89084700.0	13.8	17.2	38104200.0	353460600.0	704700.0
744.5	San Onofre 5	7314300.0	89084700.0	13.8	17.2	38104200.0	353460600.0	704700.0
744.6	San Onofre 6	4749300.0	26729100.0	9.3	16.1	30190500.0	159640200.0	550800.0
750.0	Oceanside	61629300.0	21265200.0	14.6	29.4	23031000.0	79665300.0	1741500.0
883.6	San Diego Bay	219088800.0	36895500.0	15.5	22.4	43213500.0	94916700.0	45993600.0
883.8	San Diego Bay	219088800.0	36895500.0	15.5	22.4	43213500.0	94916700.0	45993600.0
887.0	San Diego Bay	219088800.0	36895500.0	15.5	22.4	43213500.0	94916700.0	45993600.0
894.0	San Diego Bay	219088800.0	36895500.0	15.5	22.4	43213500.0	94916700.0	45993600.0
899.0	San Diego Bay	219088800.0	36895500.0	15.5	22.4	43213500.0	94916700.0	45993600.0

Table 17. Rainfall Data for Each of the 42 Mussel Watch Sites. 'RFCD' indicates the rainfall for the month of collection. 'RF1' through 'RF4' indicates the total rainfall for one to four months before collection, 'Total RF' is the total rainfall for the five months and 'AvgRF' is the average of those five months. Units are in inches of rainfall.

SN	Site Name	RF4	RF3	RF2	RF1	RFCD	Total RF	AvgRF
2.0	Crescent City/STP Outfall	11.78	5.26	3.65	9.77	6.74	37.20	7.44
3.0	Crescent City/Control	11.17	8.40	9.97	10.05	5.61	45.20	9.04
10.0	Trinidad Head	2.09	2.52	3.19	2.50	2.94	13.24	2.65
100.0	Mad River Slough	3.17	2.56	5.54	5.65	3.80	20.71	4.14
103.0	Eureka Channel	1.67	2.04	4.50	5.02	4.82	18.04	3.61
104.5	Eureka STP/Control	1.94	2.21	3.85	5.00	4.25	17.24	3.45
202.0	Bodega Head	1.46	1.35	.36	.05	.44	3.57	.73
205.0	Bodega Harbor/Spud Point Marina	2.32	4.93	6.20	5.14	2.41	20.51	4.36
336.0	J. Fitzgerald	1.10	3.29	4.56	2.96	1.59	13.49	2.70
404.0	Sandholdt Bridge	.91	1.97	2.23	3.29	1.40	9.80	1.96
414.0	Pacific Grove	2.12	3.83	3.02	3.45	2.10	14.52	2.90
430.0	Montana De Oro	.23	.35	1.25	.67	2.13	4.62	.92
434.0	Diablo Cove/South	.34	.13	1.51	.93	2.29	5.20	1.04
435.0	Intake Cove	.34	.13	1.51	.93	2.29	5.20	1.04
437.0	Point San Luis	.34	.13	1.51	.93	2.29	5.20	1.04
443.0	Diablo Cove/South/Transplant	.14	.12	1.39	.82	2.70	5.16	1.03
444.0	Intake Cove/Transplant	.30	.12	1.32	.81	2.73	5.28	1.06
445.0	San Luis Harbor/Transplant	.28	.11	1.34	.78	1.96	4.46	.89
601.0	LA Harbor/National Steel	.31	.97	.91	2.69	3.96	8.83	1.77
616.0	LA Harbor/Consolidated Slip	.28	.87	.85	2.42	3.56	7.98	1.60
618.0	LA Harbor/Angels Gate	.02	.94	2.07	1.65	4.74	9.40	1.88
648.0	Malibu	.66	.16	.78	2.48	2.55	6.62	1.32
650.0	Santa Monica	.32	.48	.55	.93	7.78	9.93	2.10
662.0	Royal Palms	.48	.65	.67	2.45	4.18	8.43	1.69
708.0	Anaheim Bay/Navy Marsh	.64	1.51	1.50	4.01	2.18	9.85	1.97
713.0	Huntington Harbour/Edinger Street	.37	.92	1.04	2.76	4.18	9.26	1.85
715.0	Huntington Harbour/Warner Ave Brdg	.33	.90	.94	2.85	4.94	9.95	1.99
723.4	Newport Bay/Turning Basin	.25	.25	.52	.62	3.84	5.47	1.09
724.0	Newport Bay/Highway 1 Bridge	.37	.89	.69	2.56	4.91	9.41	1.88
725.0	Newport Bay/Crows Nest	.33	.90	.94	2.85	4.94	9.95	1.99
726.4	Newport Bay/Rhine Channel/End	.33	.90	.93	2.85	4.94	9.95	1.99
744.1	San Onofre 1	.58	.55	.62	2.44	2.37	6.54	1.31
744.3	San Onofre 3	.77	.72	.82	2.48	2.63	6.85	1.46
744.4	San Onofre 4	.28	.71	.82	2.48	2.64	6.48	1.39
744.5	San Onofre 5	.30	.71	.82	2.48	2.63	6.48	1.39
744.6	San Onofre 6	.58	.55	.62	1.98	2.11	5.49	1.17
750.0	Oceanside	.01	.00	.26	.22	1.68	1.83	.39
883.6	San Diego Bay/7th Street Channel	.21	.30	1.03	.69	2.59	4.08	.88
883.8	San Diego Bay/Switzer Creek	.21	.66	.95	.75	2.75	4.62	.97
887.0	San Diego Bay/Evans Street	.33	.00	.76	2.62	4.30	8.01	1.95
894.0	SD Bay/Harbor Is/E Basin/Storm Dr	.06	.68	.68	1.06	5.27	7.59	1.74
899.0	San Diego Bay/Shelter Is/Fshg Pier	.50	.04	1.02	2.24	4.77	8.45	2.08

Figure 1. California Watershed Map with Mussel Watch Site Locations

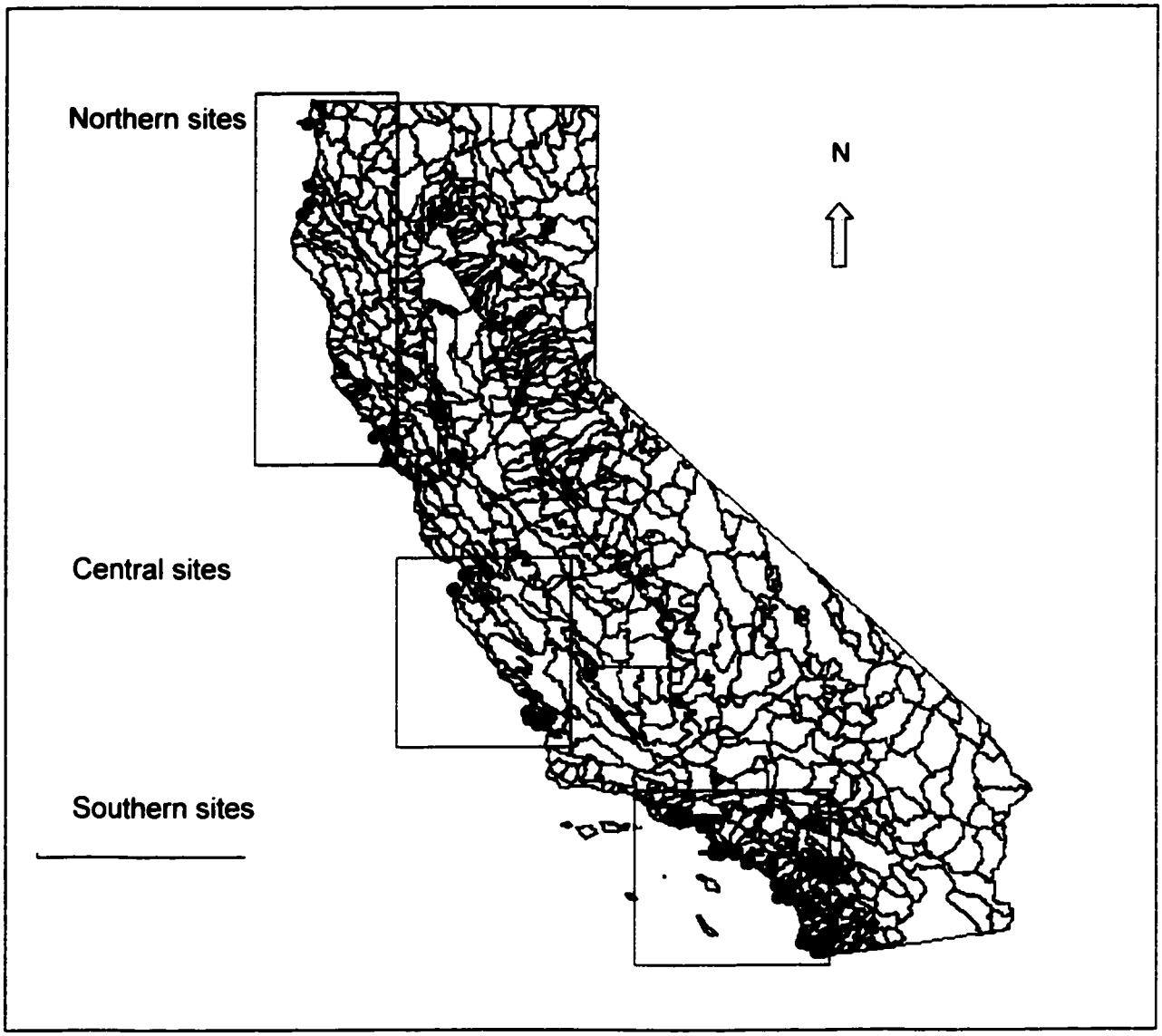

Northern sites
N
Central sites
Southern sites

Figure 2. Concentration of silver in ppm on a dry wt. basis vs. time of collection for those sites that exhibited significant linear decreasing trends.

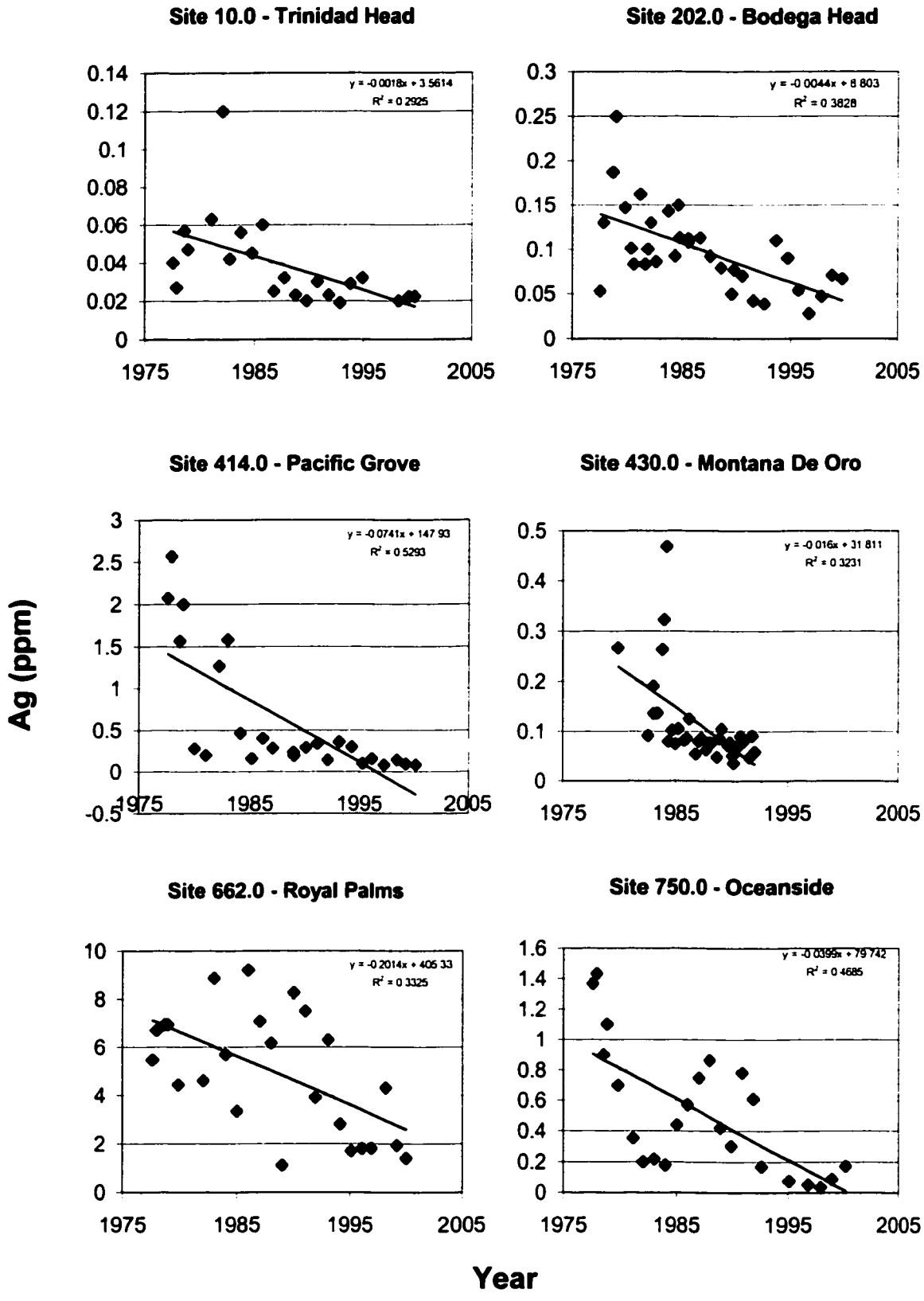

Site 10.0 - Trinidad Head
y = -0.0018x + 3.5614
R² = 0.2925
0.14
0.12
0.1
0.08
0.06
0.04
0.02
0
1975
1985
1995
2005
Site 202.0 - Bodega Head
y = -0.0044x + 8.803
R² = 0.3828
0.3
0.25
0.2
0.15
0.1
0.05
0
1975
1985
1995
2005
Site 414.0 - Pacific Grove
y = -0.0741x + 147.93
R² = 0.5293
3
2.5
2
1.5
1
0.5
0
-0.5
1975
1985
1995
2005
Site 430.0 - Montana De Oro
y = -0.016x + 31.811
R² = 0.3231
0.5
0.4
0.3
0.2
0.1
0
1975
1985
1995
2005
Site 662.0 - Royal Palms
y = -0.2014x + 405.33
R² = 0.3325
10
8
6
4
2
0
1975
1985
1995
2005
Site 750.0 - Oceanside
y = -0.0399x + 79.742
R² = 0.4685
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
1975
1985
1995
2005
Ag (ppm)
Year

Figure 3. Concentration of silver in ppm on a dry wt. basis vs. time of collection for site 894.0 that exhibited a significant non-linear decreasing trend.

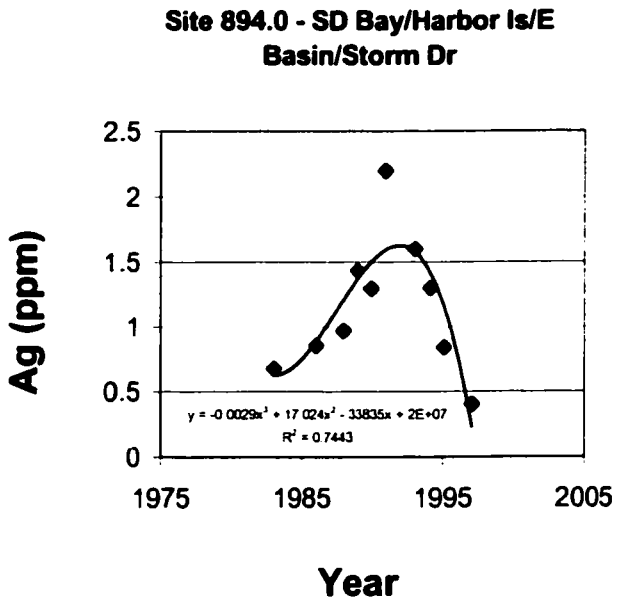

Site 894.0 - SD Bay/Harbor Is/E Basin/Storm Dr
Ag (ppm)
2.5
2
1.5
1
0.5
0
1975
1985
1995
2005
y = -0.0029x³ + 17.024x² - 33835x + 2E+07
R² = 0.7443
Year

Figure 4. Concentration of copper in ppm on a dry wt. basis vs. time of collection for those sites that exhibited significant increasing trends.

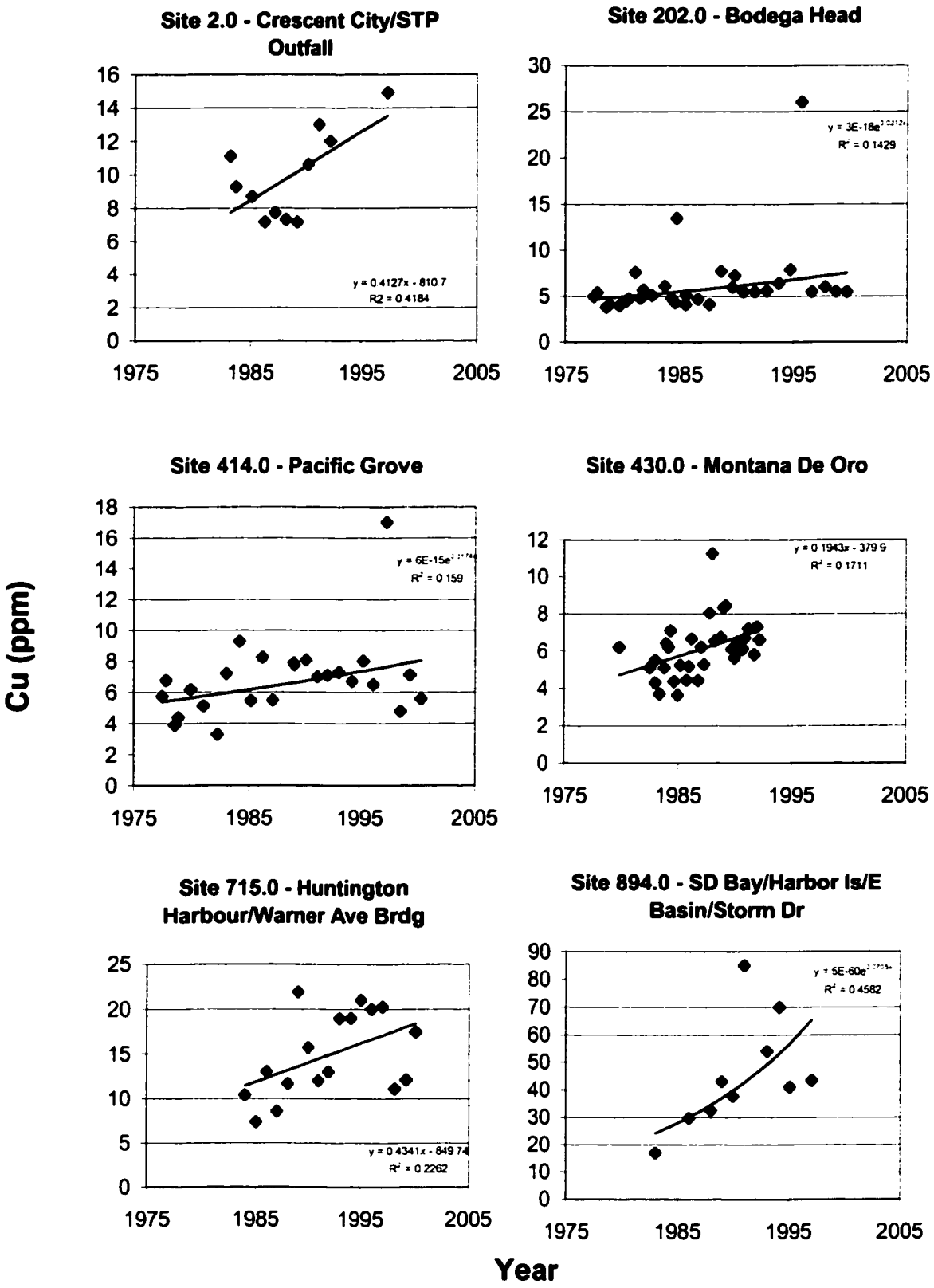

Site 2.0 - Crescent City/STP Outfall
y = 0.4127x - 810.7
R2 = 0.4184
Site 202.0 - Bodega Head
y = 3E-18e
R² = 0.1429
Site 414.0 - Pacific Grove
y = 6E-15e
R² = 0.159
Site 430.0 - Montana De Oro
y = 0.1943x - 379.9
R² = 0.1711
Site 715.0 - Huntington Harbour/Warner Ave Brdg
y = 0.4341x - 849.74
R² = 0.2262
Site 894.0 - SD Bay/Harbor Is/E Basin/Storm Dr
y = 5E-60e
R² = 0.4582
Cu (ppm)
Year

Figure 5. Concentration of copper in ppm on a dry wt. basis vs. time of collection for site 3.0 that exhibited a significant decreasing trend.

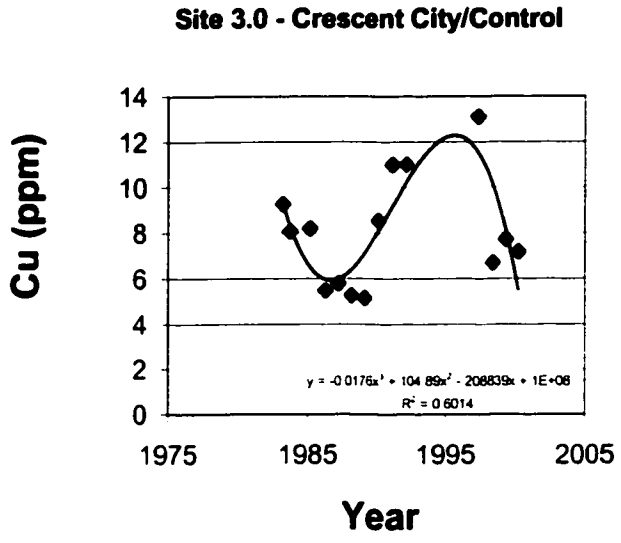

Site 3.0 - Crescent City/Control
Cu (ppm)
14
12
10
8
6
4
2
0
1975
1985
1995
2005
Year
y = -0.0176x³ + 104.89x² - 208839x + 1E+08
R² = 0.6014

Figure 6. Concentration of lead in ppm on a dry wt. basis vs. time of collection for those sites that exhibited significant decreasing trends.

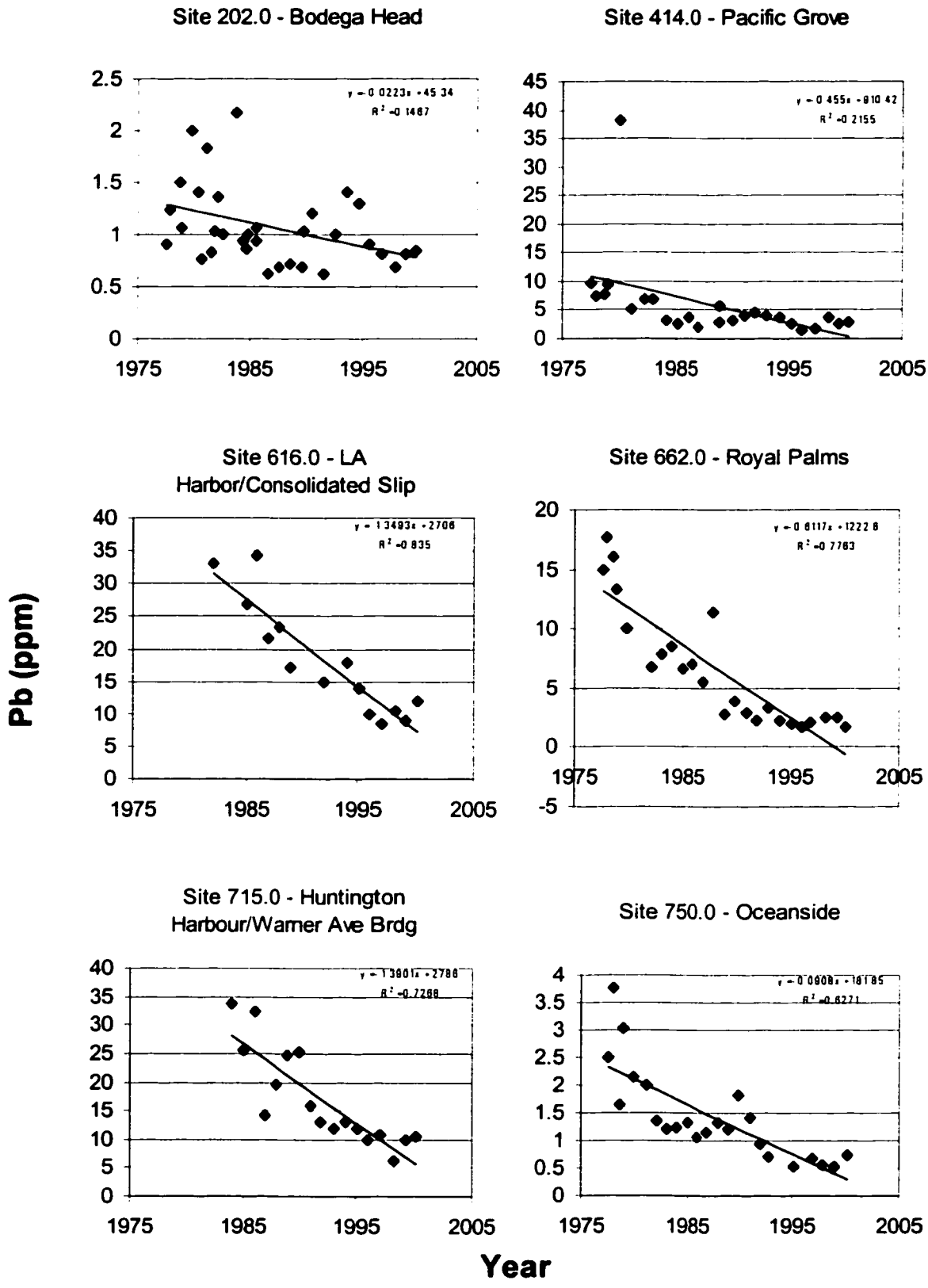

Site 202.0 - Bodega Head
Site 414.0 - Pacific Grove
Site 616.0 - LA Harbor/Consolidated Slip
Site 662.0 - Royal Palms
Site 715.0 - Huntington Harbour/Warner Ave Brdg
Site 750.0 - Oceanside
Pb (ppm)
Year
1975
1985
1995
2005

Figure 7. Concentration of total chlordane in ppb on a dry wt. basis vs. time of collection for those sites that exhibited significant decreasing trends.

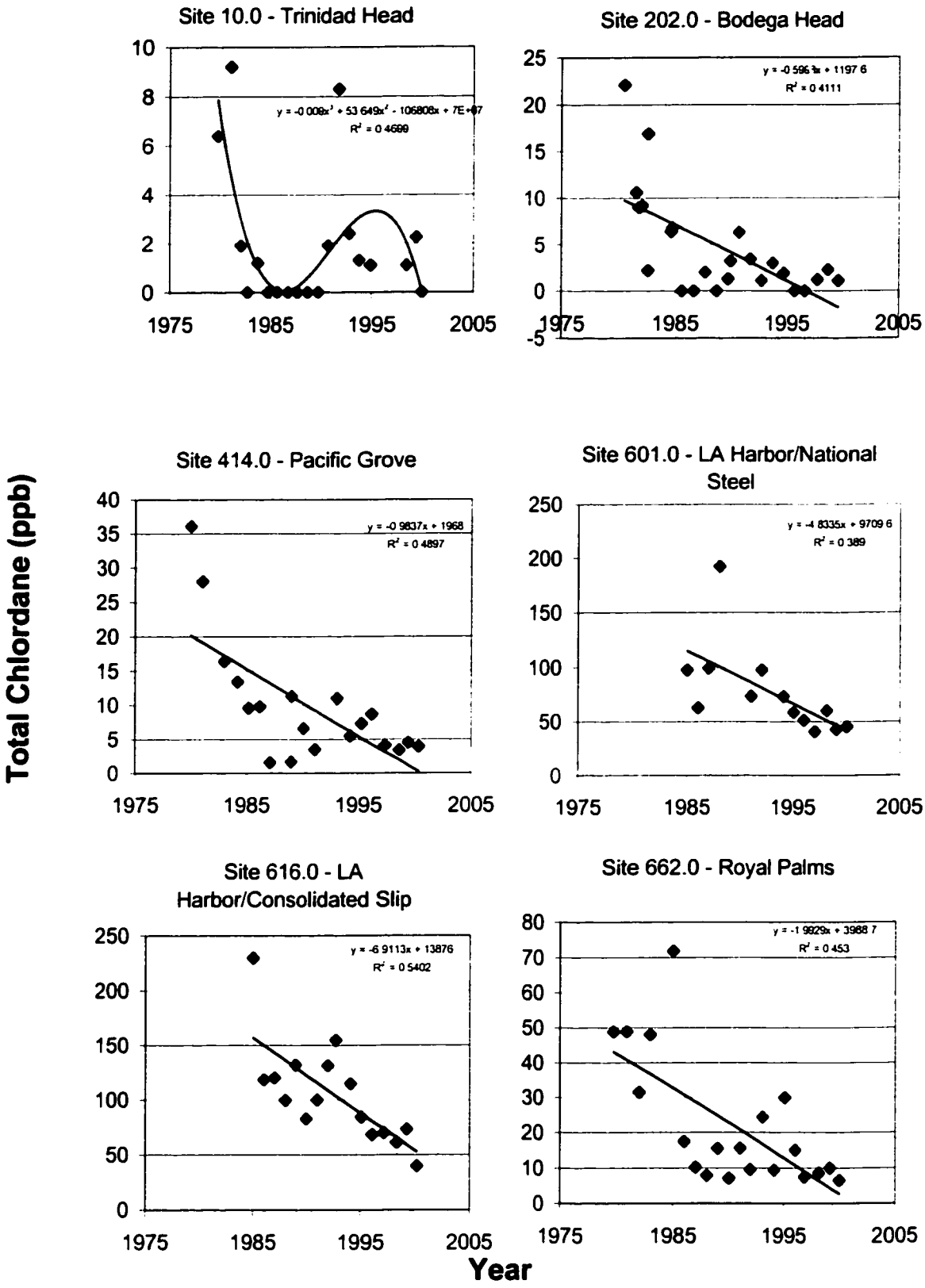

Site 10.0 - Trinidad Head
Site 202.0 - Bodega Head
Site 414.0 - Pacific Grove
Site 601.0 - LA Harbor/National Steel
Site 616.0 - LA Harbor/Consolidated Slip
Site 662.0 - Royal Palms
Total Chlordane (ppb)
Year

Figure 8. Concentration of total chlordane in ppb on a dry wt. basis vs. time of collection for those sites that exhibited significant decreasing trends.

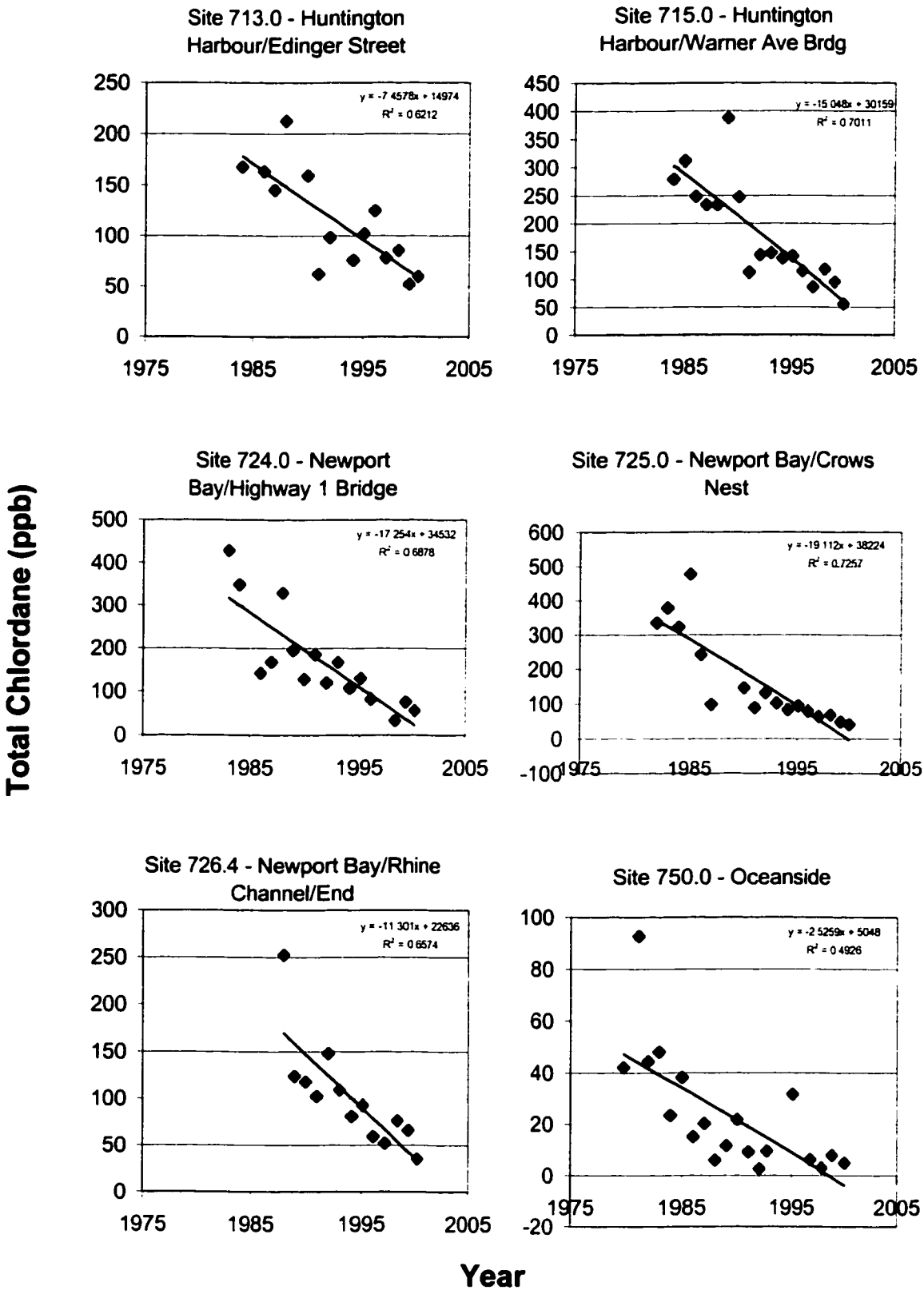

Site 713.0 - Huntington Harbour/Edinger Street
y = -7.4578x + 14974
R² = 0.6212
Site 715.0 - Huntington Harbour/Warner Ave Brdg
y = -15.048x + 30159
R² = 0.7011
Site 724.0 - Newport Bay/Highway 1 Bridge
y = -17.254x + 34532
R² = 0.6878
Site 725.0 - Newport Bay/Crows Nest
y = -19.112x + 38224
R² = 0.7257
Site 726.4 - Newport Bay/Rhine Channel/End
y = -11.301x + 22636
R² = 0.6574
Site 750.0 - Oceanside
y = -2.5259x + 5048
R² = 0.4926
Total Chlordane (ppb)
Year

Figure 9. Concentration of total chlordane in ppb on a dry wt. basis vs. time of collection for those sites that exhibited significant increasing trends.

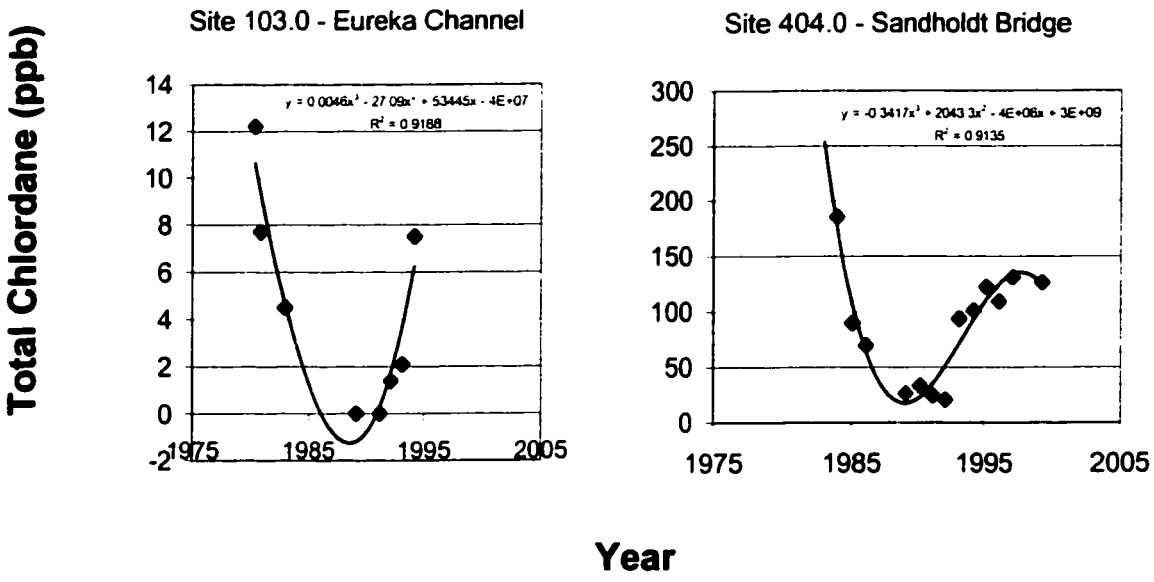
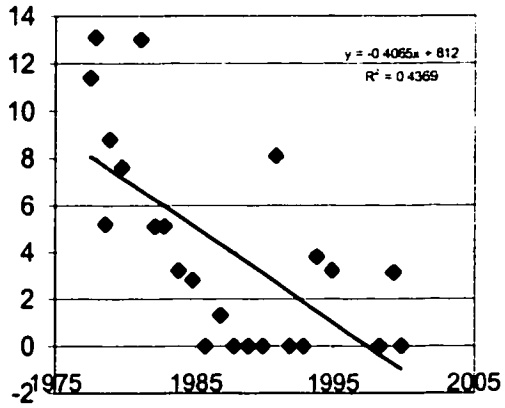
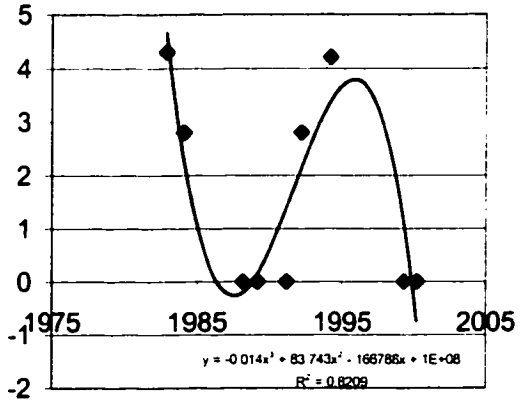

Total Chlordane (ppb)
Site 103.0 - Eureka Channel
y = 0.0046x³ - 27.09x² + 53445x - 4E+07
R² = 0.9188
14
12
10
8
6
4
2
0
-2
1975
1985
1995
2005
Site 404.0 - Sandholdt Bridge
y = -0.3417x³ + 2043.3x² - 4E+06x + 3E+09
R² = 0.9135
300
250
200
150
100
50
0
1975
1985
1995
2005
Year

Figure 10. Concentration of total DDT in ppb on a dry wt. basis vs. time of collection for those sites that exhibited significant decreasing trends.

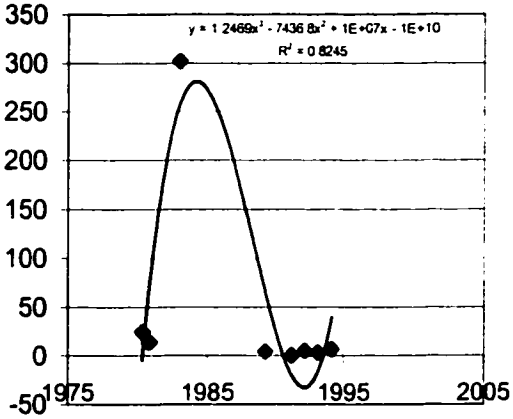
Site 10.0 - Trinidad Head



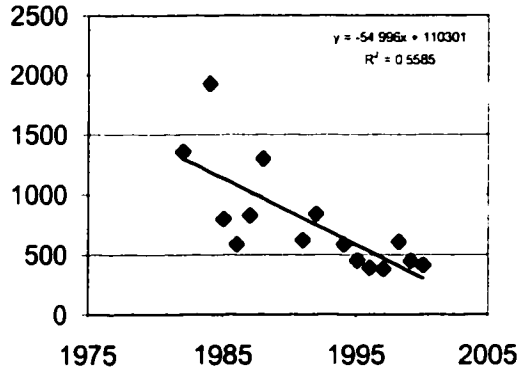
Site 100.0 - Mad River Slough



Site 103.0 - Eureka Channel

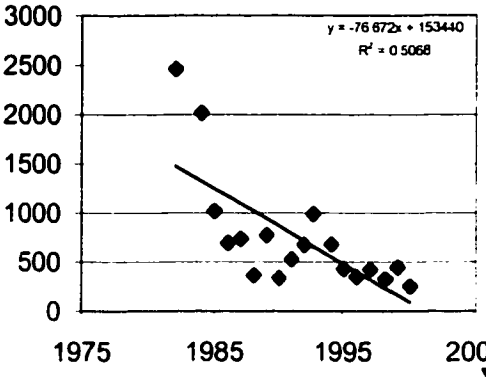


Site 601.0 - LA Harbor/National Steel

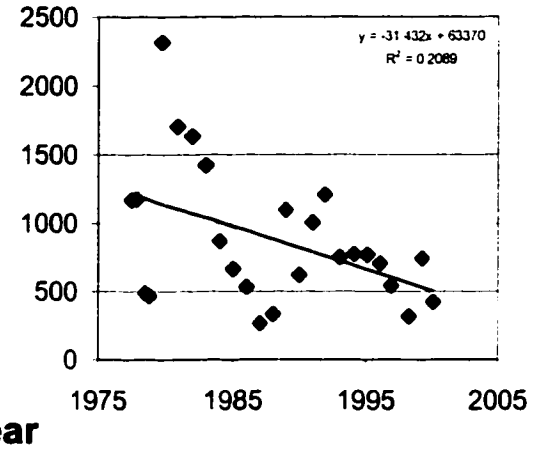


Total DDT (ppb)

Site 616.0 - LA Harbor/Consolidated Slip



Site 662.0 - Royal Palms



Year

Figure 11. Concentration of total DDT in ppb on a dry wt. basis vs. time of collection for those sites that exhibited significant decreasing trends.

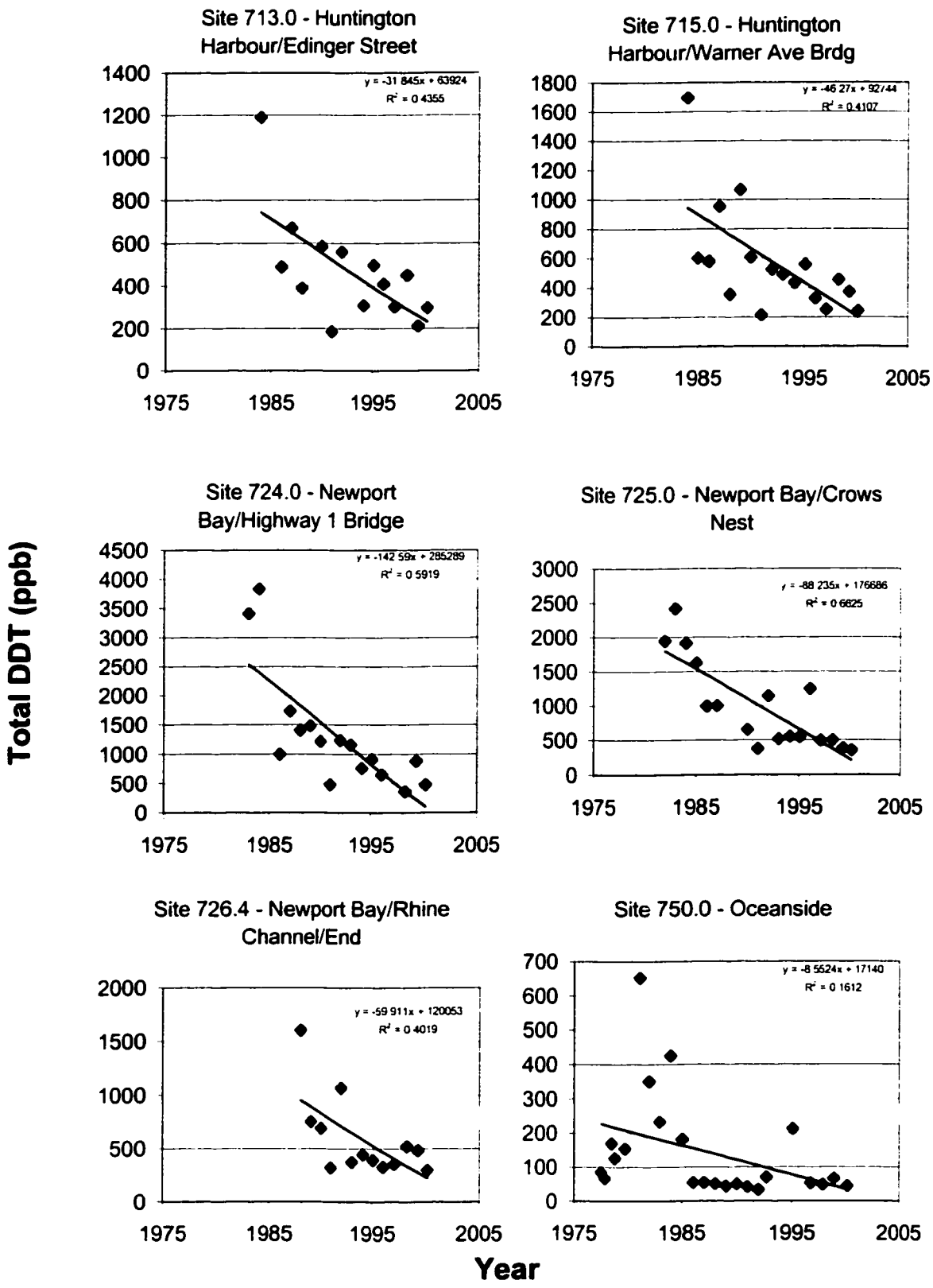

Site 713.0 - Huntington Harbour/Edinger Street
y = -31.845x + 63924
R² = 0.4355
Site 715.0 - Huntington Harbour/Warner Ave Brdg
y = -46.27x + 92744
R² = 0.4107
Site 724.0 - Newport Bay/Highway 1 Bridge
y = -142.59x + 285289
R² = 0.5919
Site 725.0 - Newport Bay/Crows Nest
y = -88.235x + 176686
R² = 0.6625
Site 726.4 - Newport Bay/Rhine Channel/End
y = -59.911x + 120053
R² = 0.4019
Site 750.0 - Oceanside
y = -8.5524x + 17140
R² = 0.1612
Total DDT (ppb)
Year

Figure 12. Concentration of total DDT in ppb on a dry wt. basis vs. time of collection for site 404.0 that exhibited a significant increasing trend.

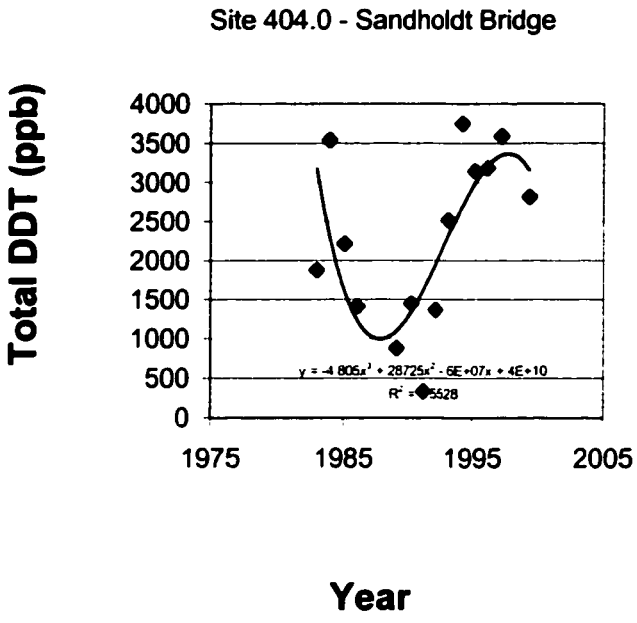

Site 404.0 - Sandholdt Bridge
Total DDT (ppb)
4000
3500
3000
2500
2000
1500
1000
500
0
1975
1985
1995
2005
Year

Figure 13. Concentration of total PCBs in ppb on a dry wt. basis vs. time of collection for those sites that exhibited significant decreasing trends.

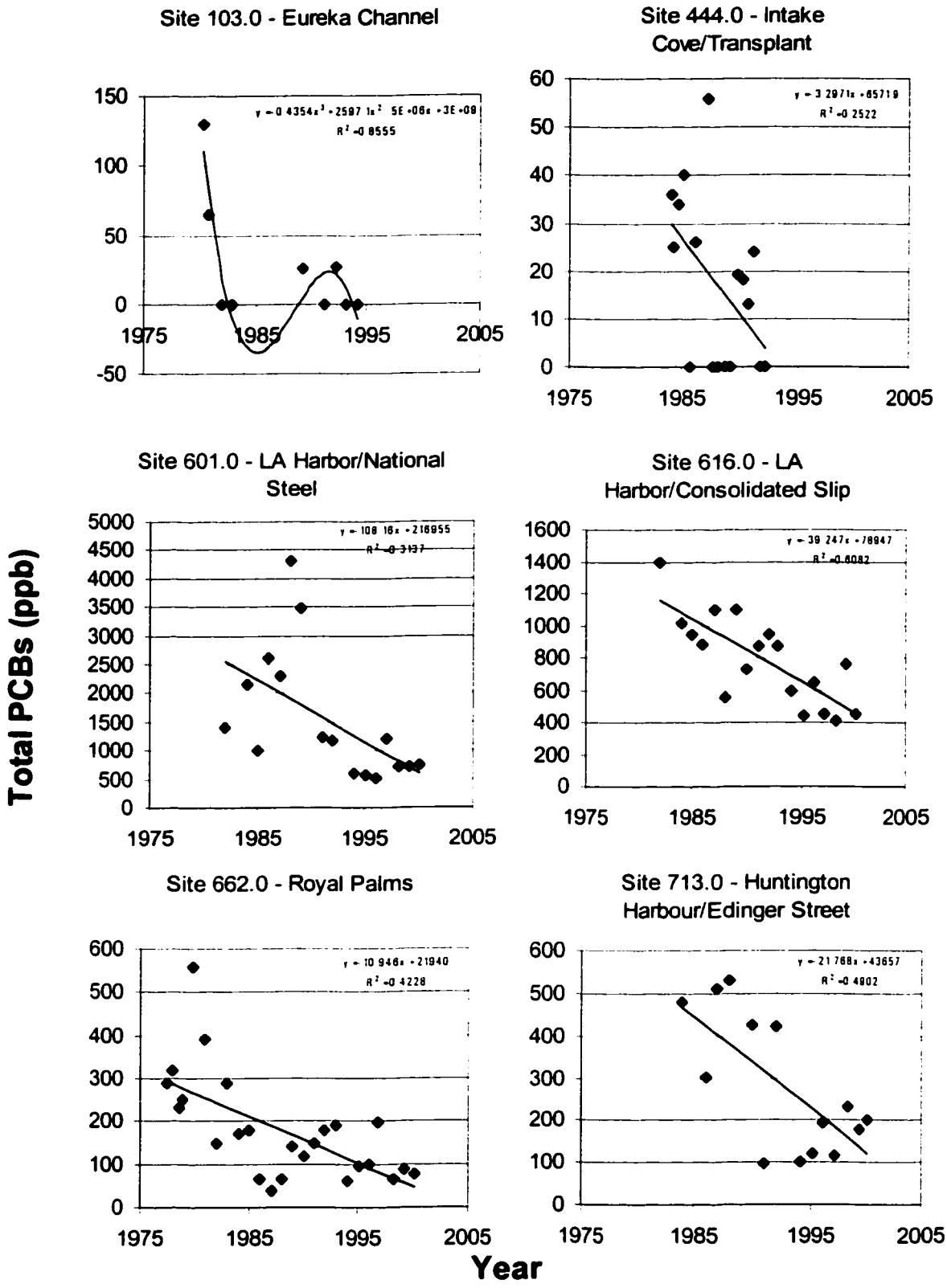

Site 103.0 - Eureka Channel
Site 444.0 - Intake Cove/Transplant
Site 601.0 - LA Harbor/National Steel
Site 616.0 - LA Harbor/Consolidated Slip
Site 662.0 - Royal Palms
Site 713.0 - Huntington Harbour/Edinger Street
Total PCBs (ppb)
Year
1975
1985
1995
2005

Figure 14. Concentration of total PCBs in ppb on a dry wt. basis vs. time of collection for those sites that exhibited significant decreasing trends.

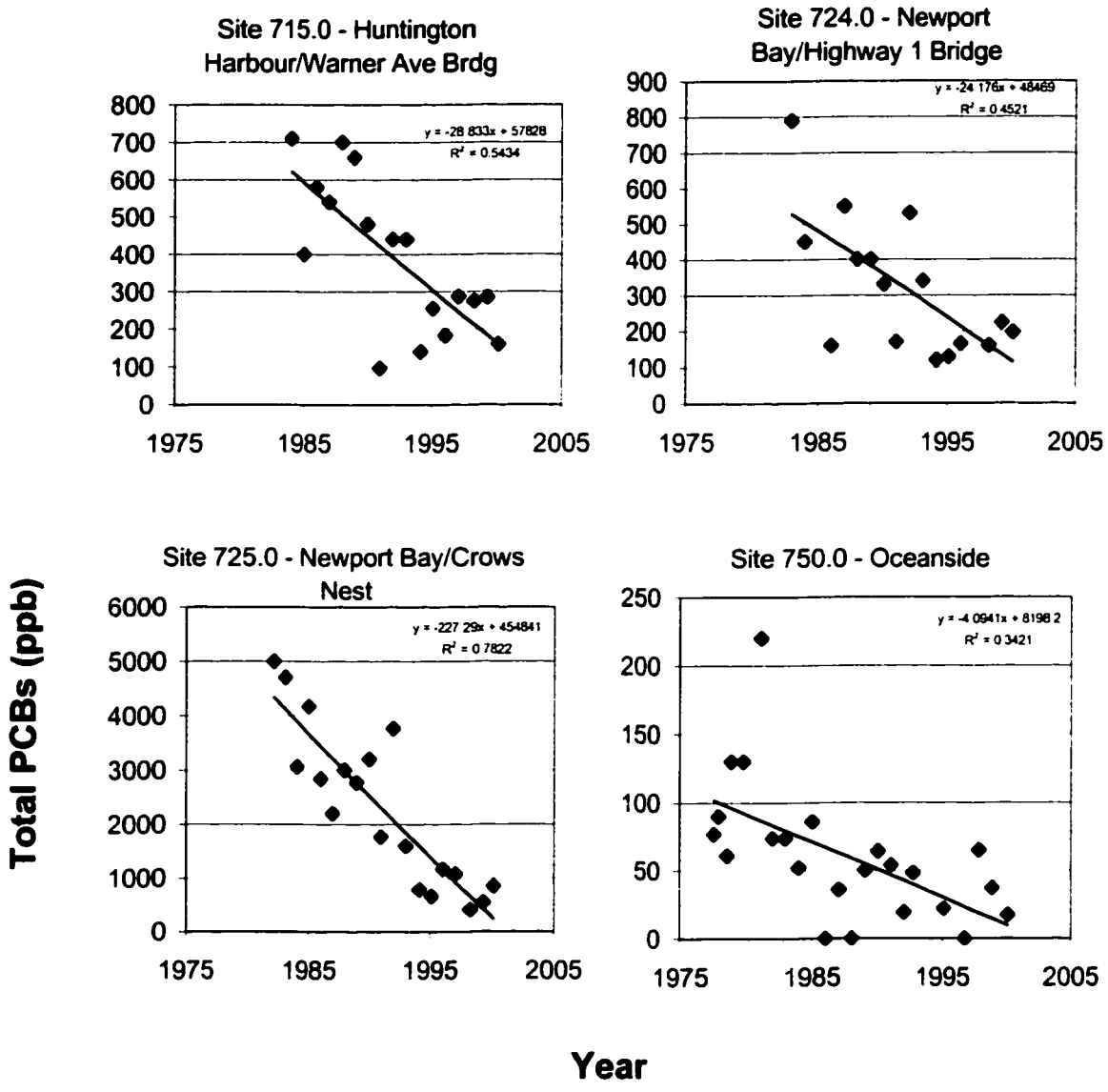

Site 715.0 - Huntington Harbour/Warner Ave Brdg
y = -28.833x + 57828
R² = 0.5434
Site 724.0 - Newport Bay/Highway 1 Bridge
y = -24.176x + 48469
R² = 0.4521
Site 725.0 - Newport Bay/Crows Nest
y = -227.29x + 454841
R² = 0.7822
Site 750.0 - Oceanside
y = -4.0941x + 8198.2
R² = 0.3421
Total PCBs (ppb)
Year

Figure 15. Concentration of total PCBs in ppb on a dry wt. basis vs. time of collection for those sites that exhibited significant increasing trends.

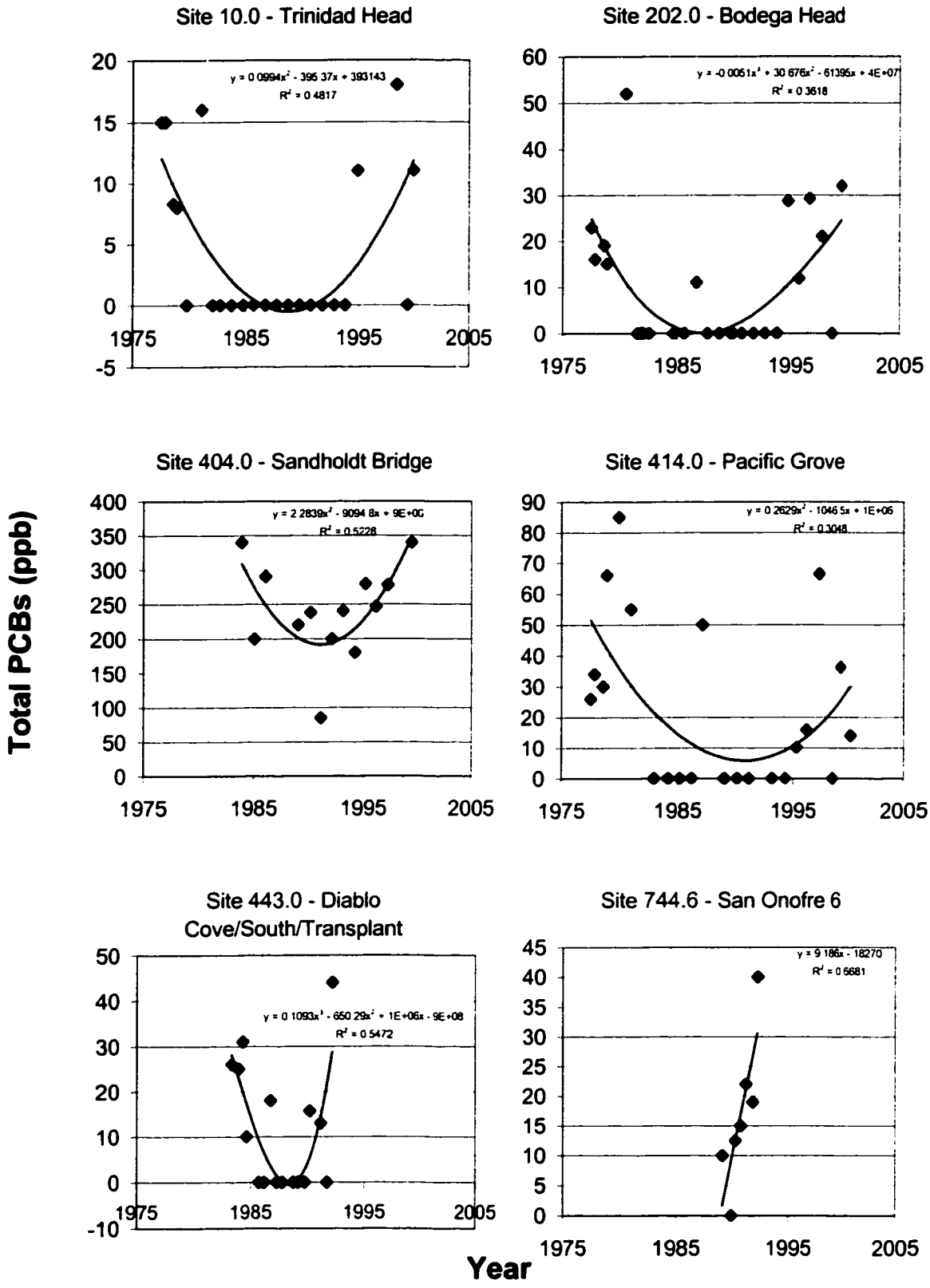

Site 10.0 - Trinidad Head
y = 0.0994x² - 395.37x + 393143
R² = 0.4817
20
15
10
5
0
-5
1975
1985
1995
2005
Site 202.0 - Bodega Head
y = -0.0051x³ + 30.676x² - 61395x + 4E+07
R² = 0.3618
60
50
40
30
20
10
0
1975
1985
1995
2005
Site 404.0 - Sandholdt Bridge
y = 2.2839x² - 9094.8x + 9E+06
R² = 0.5228
400
350
300
250
200
150
100
50
0
1975
1985
1995
2005
Site 414.0 - Pacific Grove
y = 0.2629x² - 1046.5x + 1E+06
R² = 0.3048
90
80
70
60
50
40
30
20
10
0
1975
1985
1995
2005
Site 443.0 - Diablo Cove/South/Transplant
y = 0.1093x³ - 650.29x² + 1E+06x - 9E+08
R² = 0.5472
50
40
30
20
10
0
-10
1975
1985
1995
2005
Site 744.6 - San Onofre 6
y = 9.186x - 18270
R² = 0.5681
45
40
35
30
25
20
15
10
5
0
1975
1985
1995
2005
Total PCBs (ppb)
Year

Figure 16. Concentration of total PCBs in ppb on a dry wt. basis vs. time of collection for site 894.0 that exhibited a significant non-linear increasing trend.

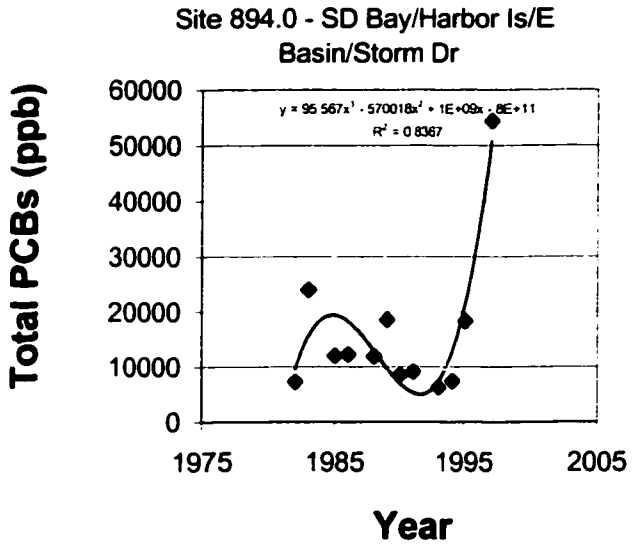
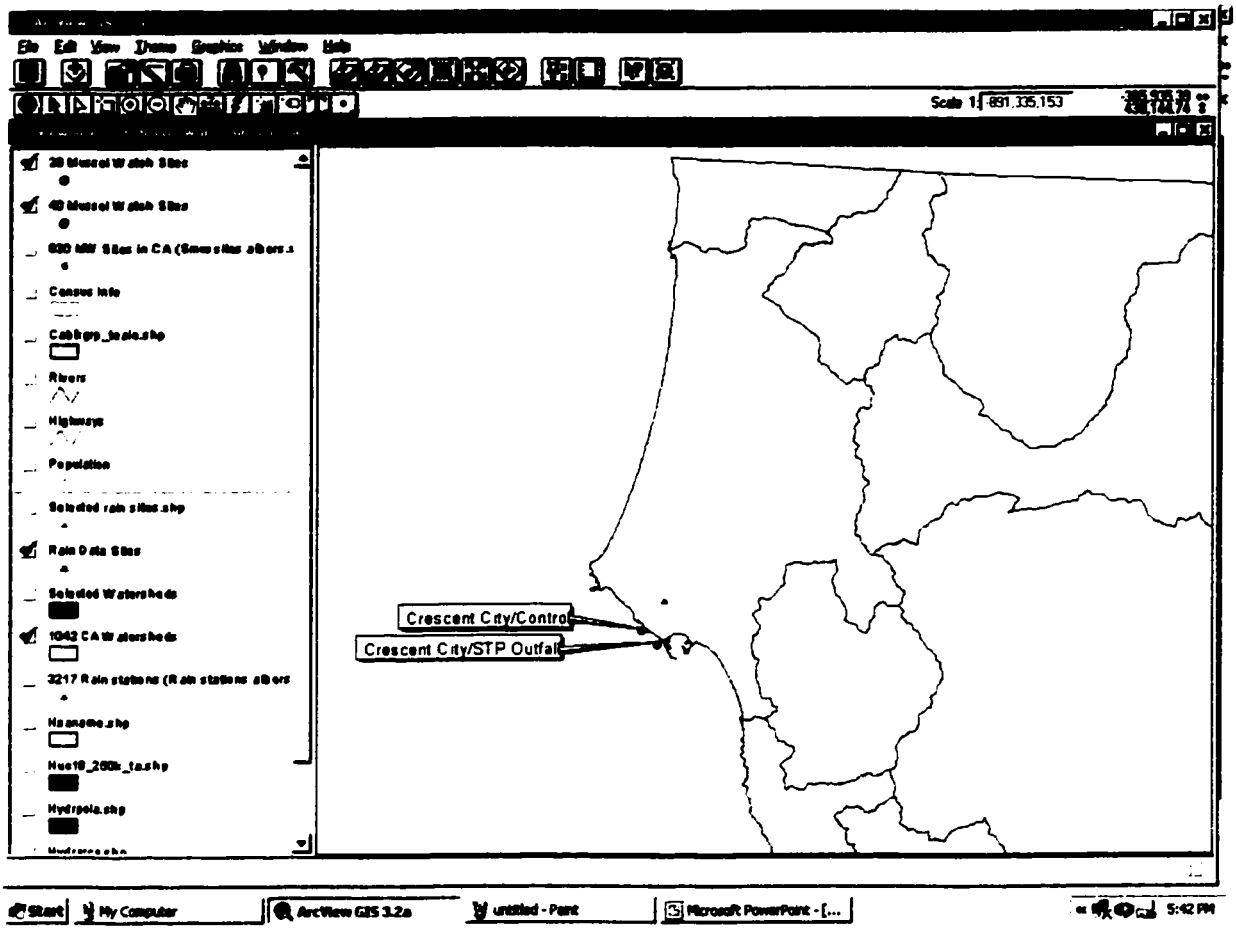

Site 894.0 - SD Bay/Harbor Is/E Basin/Storm Dr
Total PCBs (ppb)
60000
50000
40000
30000
20000
10000
0
1975
1985
1995
2005
Year

Figure 17. Example of GIS watershed map showing sites 2.0 and 3.0 in Crescent City, California.


Crescent City/Contro
Crescent City/STP Outfal

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